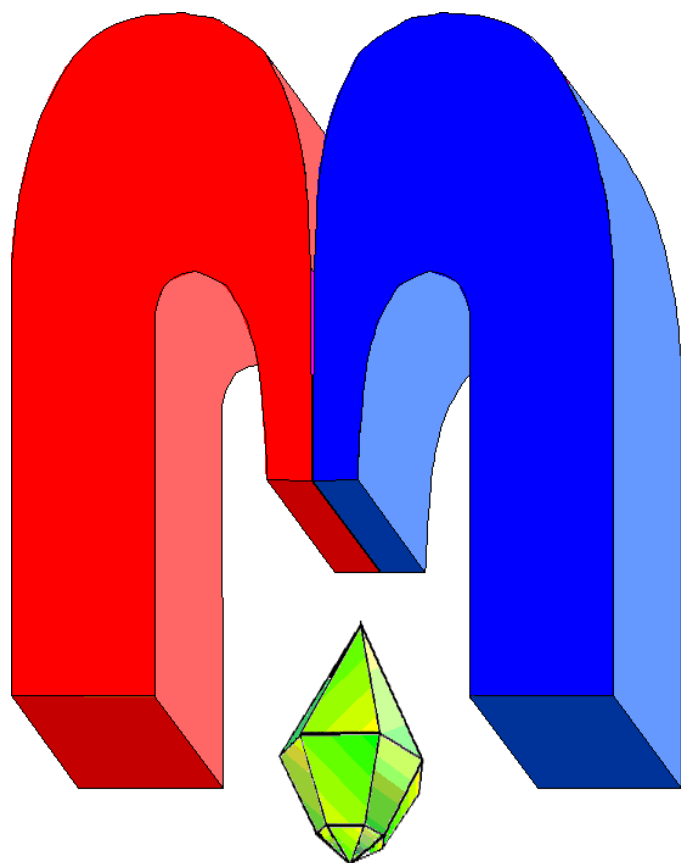


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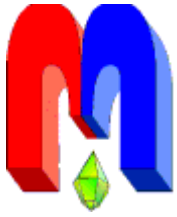
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In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Interplay of classical and quantum spin dynamics^{*}

F.S. Dzheparov

Institute for Theoretical and Experimental Physics,
B. Cheremushkinskaya, 25, Moscow 117218, Russia

E-mail: dzheparov@itep.ru

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Quantum and classical spin dynamics have more similarity than difference in theoretical part. Many processes can be described within general formalism where the type of dynamics became important at final steps only. The lecture is illustrated by consideration of operator perturbation theory and multi-spin resonance transitions.

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1. Introduction

Classical spins behavior becomes of interest in connection with modern studies of magnetic resonance in magnetic nanoparticles (see for example [1-4] and references therein).

Traditional consideration of quantum spin transitions in NMR and EPR [5, 6] was very different from analysis of ferromagnetic resonance [7], where motion of huge classical moment of total sample was used. The difference produces difficulties in comparison of corresponding results, especially for complex multi-spin and "multi-quantum" transitions.

Modern theory of multi-spin transitions starts, probably, with treating of two-spin transitions in the book [8]. Most extensive consideration was fulfilled in Ref. [9], where several versions of the theory were constructed, and intensities of many transitions were calculated and compared with precision results of beta-NMR spectroscopy [10, 11]. The theory can be characterized as a quantum mechanical unitary operator perturbation theory, constructed in commutator form. It has many common properties with treating of multi-pulse narrowing methods of solid-state NMR [12, 13]. Main aim of the lecture consists in concentrated description of the theory in application to multi-spin transitions, which is equally applicable both to quantum and classical spin systems. The description produces shortest way to separation of quantum and classical effects in results of measurements.

It should be stressed, that construction of quantum mechanical unitary operator perturbation theory started in first half of 20-th century. Probably it was initiated both internal requirements of quantum mechanics and analogies with canonical perturbation theory of classical mechanics (see Ref. [14] as an introduction for example). The influence of canonical classical mechanics was very strong from the beginning of the quantum theory. For example, quantum theory introduces new space of states instead of classical trajectories, but definition of operators in this space is strictly connected with Hamiltonian classical mechanics in Cartesian coordinates and prescribes a substitution of canonical momentum $\mathbf{p} = \partial L(\mathbf{q}, \dot{\mathbf{q}}) / \partial \dot{\mathbf{q}}$ (not kinematical one $m\dot{\mathbf{q}}$!) by the operator $\hat{\mathbf{p}} = -i\partial/\partial\mathbf{q}$ as a quantization rule. Here $L(\mathbf{q}, \dot{\mathbf{q}})$ is a Lagrangian as a function of Cartesian coordinate \mathbf{q} and velocity $\dot{\mathbf{q}}$. We will use presumably units with $\hbar=1$ and $c=1$. From mathematical point of view the quantum mechanical states form a Hilbert space, it will be referred to as Schrödinger space here in order to separate from Liouville space, discussed below.

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Interplay of classical and quantum spin dynamics

Separation of canonical and kinematical momentums gives a possibility to take into account magnetic field, because (in simplest case) the Hamiltonian

$$H(\mathbf{p}, \mathbf{q}) = \dot{\mathbf{q}}\mathbf{p} - L = \frac{1}{2}m\left(\dot{\mathbf{q}}\right)^2 + U(\mathbf{q}) = \frac{1}{2m}(\mathbf{p} + e\mathbf{A}(\mathbf{q}))^2 + U(\mathbf{q}).$$

Here $\mathbf{A}(\mathbf{q})$ is a vector-potential, e is electrical charge, and $U(\mathbf{q})$ is scalar potential energy. With more general definition we have canonical momentum as $p_\mu = \partial S(q) / \partial q^\mu$, where

$$S(q) = \int_0^q \mathbf{p}d\mathbf{q} - \int_0^t H(\mathbf{p}, \mathbf{q})dt = -\int_0^q p_\mu dq^\mu$$

is an action as a function of final 4-dimensional coordinate $q^\mu = (t, \mathbf{q})$ and integration is carried out along real trajectory. This definition produces the same space components \mathbf{p} of the 4-momentum p^μ , and new component $p^0 = -H(\mathbf{p}, \mathbf{q})$. Therefore, applying the same quantization rule to canonical momentum $p^0 = p_0 = -H(\mathbf{p}, \mathbf{q})$ we have two different definitions for corresponding momentum operator: $\hat{p}_0 = -i\frac{\partial}{\partial t}$ and $\hat{p}_0 = -H(\hat{\mathbf{p}}, \mathbf{q})$. For compatibility we should require their coincidence in action on realizable state $|\psi\rangle$, that produce the main equation of the quantum theory – Schrödinger equation $i\frac{\partial}{\partial t}|\psi\rangle = H(\hat{\mathbf{p}}, \mathbf{q})|\psi\rangle$. This short excursion demonstrates exclusive importance of canonical classical mechanics for foundations of the quantum theory.

Unitary transformations form one of the most important sections of quantum mechanics. Nowadays workers know and understand it, as a rule, much better than canonical transformations in classical theory. Similar education effect is known for a long time; it was indicated, for example, in the "Introduction" in Ref. [15]. Therefore our consideration will be based on unitary transformations in Liouville space, which can be considered as necessary and usual extension of standard Schrödinger quantum mechanical space or natural space for classical dynamics, based on Liouville equation for distribution functions. In quantum mechanics Liouville space is formed by density matrices or by usual operators of Schrödinger space.

2. Perturbation theory. General outlines

Absolute majority of interesting theoretical problems have no exact solutions. Perturbation theory produces a possibility to receive approximate (and verifiable in an experiment) result starting from exactly solvable simplified problem. From my point of view the best perturbation theory for spin dynamics consists of two very different parts. First part is directed on simplification of the Hamiltonian up to form, suitable for application in derivation of a master equation, which produces description of evolution of observables. The master equation derivation forms second part of the perturbation theory. This strategy has long history, and its elements can be found, for example, in Refs. [8] and [16].

Evolution both quantum and classic systems is governed by Liouville equation

$$\frac{\partial}{\partial t}\rho = -iL\rho, \quad (1)$$

where ρ is density matrix or distribution function in quantum and classical theory respectively. In quantum case the Liouville operator L (Liouvillian) is defined as

$$L\rho = [H, \rho] = H\rho - \rho H, \quad (2)$$

while in classical mechanics

$$L\rho = -i\{H, \rho\} = -i\left(\frac{\partial H}{\partial p} \frac{\partial \rho}{\partial q} - \frac{\partial H}{\partial q} \frac{\partial \rho}{\partial p}\right) = -i\sum_{j=1}^N \sum_{\alpha=1}^d \left(\frac{\partial H}{\partial p_j^\alpha} \frac{\partial \rho}{\partial q_j^\alpha} - \frac{\partial H}{\partial q_j^\alpha} \frac{\partial \rho}{\partial p_j^\alpha}\right). \quad (3)$$

Here N is a number of considered particles, while d is space dimension.

Eqs. (1)-(3) unify main equations of motion of quantum and classical theory. A tendency to such unification existed from the beginning of quantum mechanics. Partially one of form of main quantization postulate consists in substitution of Poisson bracket of canonical variables by the operator commutator according the same rule $\{p_j^\alpha, q_k^\beta\} = \delta_{jk} \delta_{\alpha\beta} \rightarrow i\left[\hat{p}_j^\alpha, \hat{q}_k^\beta\right] = \delta_{jk} \delta_{\alpha\beta}$. Heisenberg equations of motion have the same similarity to Hamilton equations.

From operator point of view the Liouvillian L is a Hermite operator in a new (relative to Schrödinger space) Hilbert space (Liouville space), where density matrices and other quantum mechanical operators works as vectors, while in classical theory Liouville space is formed by distribution functions and other functions in the same way. The Liouville operator is hermitian relative to scalar production $(a, b) = \text{Tr}(a^+ b)$ or $(a, b) = \int dpdq \cdot a^+ b$ in quantum and classical theory respectively.

Formal Nakajima-Zwanzig derivation of master equation starts from separation of a small, but important part ρ_1 of ρ , which is sufficient for calculation of observables. In simplest case this operation is introduced by a time-independent projection operator P and

$$\rho_1 = P\rho, \quad P^2 = P. \quad (4)$$

Multiplying Eq. (1) on P we have

$$\frac{\partial}{\partial t} \rho_1 = \frac{\partial}{\partial t} P\rho = -iPL(P + \bar{P})\rho = -i(PLP\rho_1 + PL\bar{P}\rho_2), \quad (5)$$

where

$$\rho_2 = \bar{P}\rho, \quad \bar{P} = 1 - P, \quad (\bar{P})^2 = \bar{P}, \quad (6)$$

and, evidently,

$$\frac{\partial}{\partial t} \rho_2 = -i\bar{P}L(P + \bar{P})\rho = -i(\bar{P}LP\rho_1 + \bar{P}L\bar{P}\rho_2). \quad (7)$$

Solving Eq. (7) with initial condition $\rho_2(t=0) = 0$ and substituting the solution into Eq. (5) we receive a master equation

$$\frac{\partial}{\partial t} \rho_1 = -i\Omega\rho_1 - \int_0^t d\tau M(t, \tau)\rho_1(\tau) \quad (8)$$

with definitions of frequency matrix Ω and memory kernel $M(t, \tau)$:

$$\Omega = PLP, \quad M(t, \tau) = PL(t)\bar{P} \cdot T \exp\left(-i\int_\tau^t d\tau_1 \bar{P}L(\tau_1)\bar{P}\right) \cdot \bar{P}L(\tau)P. \quad (9)$$

Here standard chronological exponent ($T\exp$) is introduced.

These formal operations are well known and equally applicable both for quantum and classical theory. Real calculations require reasonable approximations for memory kernel which can depend on type of dynamics and we will not discuss them later.

Interplay of classical and quantum spin dynamics

We will concentrate our attention on the first part of perturbation theory consisting of simplification of the Hamiltonian. We will consider the Hamiltonian of the form

$$H = \sum_m H_m e^{i\omega_m t}, \quad H_m^+ = H_{-m}. \quad (10)$$

As a rule the representation (10) is a consequence of application of so called representation of interaction, when strong but exactly solvable part of the evolution is excluded from the equation of motion, and all terms in (10) have the same order of value, while many of frequencies ω_m are large.

We come to this representation, for example, in discussion of resonances at frequencies, obeying the condition $k\omega = m\omega_l$. Here k and m are integer, and ω_l and ω are Larmor frequency and frequency of alternating field. The transitions can take place, for example, in homo-spin system formed by spins I , placed in strong static magnetic field (directed along z -axis with value ω_l) and orthogonal it radio-frequency field with value ω_l (we use frequency units for magnetic fields) in presence of dipole-dipole interactions. The terms H_m at that represent so called Van Vleck' alphabet [5, 7] (with corresponding frequencies $\{\omega_m\} = 0, \pm\omega_l, \pm 2\omega_l$) and rf-interaction (with frequencies $\pm(\omega_l - \omega)$) in the system, rotating with the frequency ω_l around the static field. Other example is produced by nuclear or electron spins in presence of quadrupole interaction

$$H_Q = \omega_Q \left((\mathbf{nI})^2 - \frac{1}{3} I(I+1) \right) \quad (11)$$

and static magnetic field, if $\omega_Q \ll \omega_l$. The Hamiltonian (11) produces the same set of primary frequencies ω_m as homo-spin dipole interaction. More complex examples together with solutions can be found in Refs. [9-11].

The aim of considered perturbation theory consists in such transformation of the Liouville equation, which conserve Hamilton form (2) of the main equation (with a new Hamiltonian) and suppress fast oscillating terms of the Hamiltonian. Slow oscillating terms should be treated via master equation.

Very important property of the theory consists in the fact, that new (transformed) Hamiltonian is constructed from powers of commutations (or Poisson brackets) that automatically produce no volume divergences.

Other important property is specific for spin dynamics, where spin variables are included only instead of full set of coordinates and momenta, and main quantum mechanical (QM) commutators are in exact agreement with Poisson brackets of classical mechanics (CM) again:

$$[I_j^\alpha, I_k^\beta] = i\delta_{jk} e_{\alpha\beta\gamma} I_k^\gamma. \quad (12)$$

Here I_j^α is α component of j -th spin, and $[a, b]$ means the commutator of operators a and b in quantum mechanics, or $[a, b] = -i\{a, b\}$ in classical theory. A summation is meant in (12) over index γ on right side, which is absent on left side. With these notations Eqs. (1)-(3) can be written as

$$\frac{\partial}{\partial t} \rho = -iL\rho = -i[H, \rho] = -iH^\times \rho \quad (13)$$

both in quantum and in classical theory, and the operation $[a, b]$ will be referred as commutator in both theories, if it will not require additional refinement. Last relation in (13) can be written as $L = H^\times$, and it indicates that operator L in Liouville space (superoperator) is formed by H via commutator that is rather special form for superoperators.

3. Unitary operator perturbation theory

We can separate the Hamiltonian (10) (and corresponding Liouvillian) into fast (F) and slow (S) oscillating parts:

$$H(t) = H_S(t) + H_F(t), \quad H_S(t) = \sum_{|\omega_m| < \Omega} e^{i\omega_m t} H_m, \quad H_F(t) = \sum_{|\omega_m| > \Omega} e^{i\omega_m t} H_m. \quad (14)$$

The boundary frequency Ω should be defined later from the requirement of self-consistency of calculations.

We can introduce a new variable $\rho^{(1)}(t)$ via unitary transformation in Liouville space

$$\rho(t) = U(t)\rho^{(1)}(t) = \exp(-iS^\times(t))\rho^{(1)}(t), \quad S^\times(t) = \sum_{|\omega_m| > \Omega} \frac{1}{i\omega_m} e^{i\omega_m t} H_m^\times. \quad (15)$$

In action on typical states, for example on I_j^α or $I_j^\alpha I_k^\beta$, the superoperator H_m^\times produces finite result even for infinite systems, when number of spins $N \rightarrow \infty$. Therefore

$$S^\times(t) \sim H_F^\times / \Omega \sim \varepsilon \quad (16)$$

can be considered as small value proportional to small parameter ε .

It is evident that $\partial S^\times(t) / \partial t = H_F^\times(t)$, and equation of motion for $\rho^{(1)}(t)$ does not contain fast oscillating terms in main order in ε :

$$\frac{\partial}{\partial t} \rho^{(1)} = -iH_S^\times \rho^{(1)} + O(\varepsilon). \quad (17)$$

More exactly

$$\frac{\partial}{\partial t} \rho^{(1)} = -iL^{(1)} \rho^{(1)}, \quad L^{(1)} = U^+ L U + i\dot{U}^+ U. \quad (18)$$

It is evident, that

$$U^+ L U \rho^{(1)} = U^+ \left[H, (U \rho^{(1)})_0 \right] = \left[(U^+ H)_0, \rho^{(1)} \right]. \quad (19)$$

Here a symbol $(U \rho^{(1)})_0$ (or $(U^+ H)_0$) means, that action of superoperators U (or U^+) is concentrated within the bracket, and the result is a vector in Liouville space.

Differentiation of the exponential operator $U(t)$ is a standard action (see for example [9] or Appendix in [17]) with a result

$$\frac{\partial}{\partial t} e^{iS^\times} = i \int_0^1 d\alpha e^{i\alpha S^\times} \dot{S}^\times e^{i(1-\alpha)S^\times}. \quad (20)$$

Therefore

$$\begin{aligned} \dot{U}^+ U \rho^{(1)} &= i \int_0^1 d\alpha e^{i\alpha S^\times} \dot{S}^\times e^{-i\alpha S^\times} \rho^{(1)} = i \int_0^1 d\alpha e^{i\alpha S^\times} H_F^\times e^{-i\alpha S^\times} \rho^{(1)} \\ &= i \int_0^1 d\alpha e^{i\alpha S^\times} \left[H_F, (e^{-i\alpha S^\times} \rho^{(1)})_0 \right] = i \int_0^1 d\alpha \left[(e^{i\alpha S^\times} H_F)_0, \rho^{(1)} \right] = i \left[\left(\frac{e^{iS^\times} - 1}{iS^\times} H_F \right)_0, \rho^{(1)} \right]. \end{aligned} \quad (21)$$

We see that new Liouvillian $L^{(1)} = H^{(1)\times}$ is formed of new Hamiltonian according to relations (18), (19) and (21) as

$$L^{(1)} \rho^{(1)} = H^{(1)\times} \rho^{(1)} = [H^{(1)}, \rho^{(1)}], \quad H^{(1)} = e^{iS^\times} H - \frac{e^{iS^\times} - 1}{iS^\times} H_F. \quad (22)$$

The transformation from Eqs. (13) and (14) to (18) and (22) defines an iteration method, which can be continued later. At every step of iterations fast oscillating term of the Hamiltonian is suppressed, and new slow oscillating terms are created. After n steps the fast oscillating term has an order $H_F^{(n)} \sim \varepsilon^{2^n}$ as in super-convergent classical Kolmogorov-Arnol'd-Moser theory. But, for typical conditions, relaxation speed $w_F^{(n)}$, produced by $H_F^{(n)}$, is not smaller than the speed $w_F = w_F^{(n=0)}$, calculated directly from $H_F \sim \varepsilon^0$ [8, 9]. There is no contradiction here, because, as a rule, $|\ln w_F^n| \sim 1/\varepsilon$, while the iterations display the power accuracy $\sim (\varepsilon^{2^n})^2$ for $w_F^{(n)}$ only [9].

Main result for applications is concentrated in effective Hamiltonian of slow motions H_S^{eff} presented by slow part of the new Hamiltonian, because it contains new terms relative to initial H_S . Two iterations produce accuracy up to ε^3 and [9]

$$H_S^{eff} = H_S^{(2)} + O(\varepsilon^4) = \left(\left(1 - \frac{1}{2} S^{\times 2} - \frac{i}{6} S^{\times 3} \right) H_S + \left(\frac{i}{2} S^\times - \frac{1}{3} S^{\times 2} - \frac{i}{8} S^{\times 3} \right) H_F \right)_S - \frac{i}{2} \left[\left[\overline{\left(S^\times \left(H_S + \frac{1}{2} H_F \right) \right)}_{0F}, \left(S^\times \left(H_S + \frac{1}{2} H_F \right) \right)_{0F} \right] \right]_S + O(\varepsilon^4). \quad (23)$$

Here $\tilde{A} = \sum_m A_m \exp(i\omega_m t) / (i\omega_m)$ for $A = \sum_m A_m \exp(i\omega_m t)$, the symbol $(\dots)_0$ is introduced in (19), and subscripts S and F indicate separation of slow and fast oscillating parts, as in (14). Main attention of many workers was attracted to situations, where instead of separation of slow part of the Hamiltonian the time averaging can be used. Corresponding result with accuracy up to ε^2 was received in Refs. [18, 19], it was repeated for spin dynamics in [13], and the accuracy was refined up to ε^5 in Ref. [20].

4. An example

Let us consider a resonance at the frequency $\omega = 2\omega_I$ in homo-spin system, produced by dipole interactions as an example. The Hamiltonian can be written as

$$H = H_Z + H_D + H_{rf}(t). \quad (24)$$

Here $H_Z = \omega_I I_z$ is Zeeman interaction, H_D is dipole-dipole Hamiltonian and

$$H_{rf}(t) = \frac{1}{2} \omega_I \left(I_+ e^{-i\omega t} + I_- e^{i\omega t} \right) \quad (25)$$

represents the action of resonance alternating field. Main influence on resonance at the frequency $\omega = 2\omega_I$ is produced by so called C-term of H_D :

$$H_{DC} = \frac{1}{2} \sum_{jk} \left(c_{jk} \left(I_j^z I_k^+ + I_j^+ I_k^z \right) + H.C. \right) = H_C^+ + H_C^-, \quad (26)$$

$$c_{jk} = -\frac{3}{4} \frac{\gamma^2 \hbar^2}{r_{jk}^3} \sin(2\mathcal{G}_{jk}) e^{i\phi_{jk}}, \quad [H_Z, H_C^\pm] = \pm \omega_I H_C^\pm,$$

where polar angles \mathcal{G}_{jk} and ϕ_{jk} define direction of the interspin vector \mathbf{r}_{jk} .

Effective Hamiltonian of slow motion will include secular part of dipole interactions H_{D0} and, according to (22), a resonance term

$$H_{res}(t) = \frac{i}{2} [S, H_F]_S, \quad H_F = H_C^+ e^{i\omega_I t} + H_C^- e^{-i\omega_I t} + \frac{1}{2} \omega_{I1} \left(I_+ e^{i(\omega_I - \omega)t} + I_- e^{-i(\omega_I - \omega)t} \right),$$

$$S = \widetilde{H}_F = H_C^+ \frac{e^{i\omega_I t}}{i\omega_I} - H_C^- \frac{e^{-i\omega_I t}}{i\omega_I} + \frac{\omega_{I1}}{2i(\omega_I - \omega)} \left(I_+ e^{i(\omega_I - \omega)t} - I_- e^{-i(\omega_I - \omega)t} \right).$$
(27)

After simplifying

$$H_{res}(t) = \frac{\omega_{I1} \omega}{4\omega_I (\omega - \omega_I)} e^{i(2\omega_I - \omega)t} \sum_{jk} c_{jk} I_j^+ I_k^+ + H.C. = H_r^+(t) + H_r^-(t),$$

$$H_r^+(t) \approx \frac{\omega_{I1}}{2\omega_I} e^{i(2\omega_I - \omega)t} \sum_{jk} c_{jk} I_j^+ I_k^+, \quad H_r^-(t) = (H_r^+(t))^+.$$
(28)

In the simplest theory (neglecting the influence of dipole order) important part of the density matrix can be chosen as a function of I_z only and master equation (8) after standard transformations became a form

$$\frac{\partial}{\partial t} \rho_1 = - \int_0^\infty d\tau \left[H_{res}(t, t), [H_{res}(t - \tau, t - \tau), \rho_1(t)] \right],$$

$$H_{res}(t, \tau) = e^{iH_{D0}t} H_{res}(\tau) e^{iH_{D0}\tau}.$$
(29)

Correspondingly

$$\frac{\partial}{\partial t} \langle I_z \rangle = \frac{\partial}{\partial t} (I_z, \rho_1) = - \int_0^\infty d\tau \left(I_z, [H_{res}(t, t), [H_{res}(t - \tau, t - \tau), \rho_1(t)]] \right)$$

$$= -2 \int_{-\infty}^\infty d\tau e^{i(2\omega_I - \omega)\tau} \left([H_r^+(\tau, 0), H_r^-], \rho_1 \right).$$
(30)

Here scalar production in Liouville space is applied and the relation $[H_z, H_r^\pm] = \pm 2\omega_I H_r^\pm$ is taken into account.

One of most representative parameter of resonance at combination frequency $\omega = k\omega_I$ is its forbidding factor $A(\omega = k\omega_I)$. To define it we can introduce a measurable parameter

$$W_0(\omega = 2\omega_I) = - \lim_{t \rightarrow 0} \int d\omega \frac{\partial}{\partial t} \ln \langle I_z \rangle = \frac{4\pi \langle [H_r^+, H_r^-] \rangle_0}{\langle I_z \rangle_0},$$
(31)

which presents intensity of the resonance. Here $\langle F \rangle_0 = (F, \rho_1(t=0))$ for any $F = F^+$. The argument $\omega = 2\omega_I$ on the left side in (30) indicates type of the resonance, while frequency integration in second term is fulfilled near the resonance frequency $2\omega_I$ and does not include resonances of other types, which are supposed as well separable. The limit $t \rightarrow 0$ in (31) implies small $t \gg T_2$, according to applicability of Eq. (29). Similar parameter for Larmor resonance is

$$W_0(\omega = \omega_I) = \pi \omega_{I1}^2.$$
(32)

The forbidding factor is

$$A(\omega = 2\omega_I) = W_0(\omega = 2\omega_I) / W_0(\omega = \omega_I).$$
(33)

We see that it depends on equilibrium properties of the system. Of course results will be very different for large classical spins and for small quantum spins with the same gyromagnetic ratio. Direct calculation produces

$$W_0(\omega = 2\omega_l) = 8\pi \left(\frac{\omega_{l1}}{\omega_l} \right)^2 \frac{1}{N} \sum_{jk} |c_{jk}|^2 \left\langle I(I+1) - (I_k^z)^2 \right\rangle. \quad (34)$$

It is evident that $W_0(\omega = 2\omega_l)$ does not depend on spin value at all. Contrary that $W_0(\omega = 2\omega_l)$ has strong corresponding dependence, and "extra"-quantum case $I = 1/2$ has no dependence on initial state, while classic limit has strong corresponding dependence.

5. Conclusion

The analysis indicates that description of classical and quantum spin dynamics can be carried out in general way, where difference between these theories go into action at last steps in calculations of such values as forbidding factor or resonance form function for example. These exist strong indications, that dipole resonance form function for classical and quantum theories are very close as well, see for example [21].

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