

## Research Article

# Quantum Field Theory of Dynamics of Spectroscopic Transitions by Strong Dipole-Photon and Dipole-Phonon Coupling

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Matrix-operator difference-differential equations for dynamics of spectroscopic transitions in 1D multiqubit exchange-coupled (para)magnetic and optical systems by strong dipole-photon and dipole-phonon coupling are derived within the framework of quantum field theory. It has been established that by strong dipole-photon and dipole-phonon coupling the formation of long-lived coherent system of the resonance phonons takes place, and relaxation processes acquire pure quantum character. It is determined by the appearance of coherent emission process of EM-field energy, for which the resonance phonon system is responsible. Emission process is accompanied by phonon Rabi quantum oscillation, which can be time-shared from photon quantum Rabi oscillations, accompanying coherent absorption process of EM-field energy. For the case of radio spectroscopy, it corresponds to the possibility of the simultaneous observation along with (para)magnetic spin resonance, the acoustic spin resonance.

## 1. Introduction

The use of optical and radio spectroscopy methods to create coherent states in solid materials has numerous potential applications, ranging from low-power nonlinear optics to high-temperature spectral hole burning memories to solid-state quantum computing. The interest in optical excitation lies in the fact that the coherent states can be efficiently excited and manipulated using optical laser fields, yet they are weakly coupled to the environment and hence have the long coherence lifetimes needed for optical memories and quantum computing.

It seems to be very substantial for practical applications and even necessary the development of the theory, which allows to predict the appropriate electronic systems and the conditions for the formation of long-lived coherent states. Subsequent progress in given field seems to be connected with the elaboration of theoretical models based on quantum field theory (QFT) including quantum electrodynamics (QED). Really, quantum field theory including quantum electrodynamics, in fact, becomes to be working instrument

in spectroscopy studies and industrial spectroscopy control. Moreover, we will show, in given report, that quantized electromagnetic (EM) field itself and quantized field of lattice deformations (phonon field) will be in the nearest future the working components of optoelectronic, spintronic devices and various logic quantum systems including quantum computers and quantum communication systems.

Quantum dynamics of two-level systems (qubits), coupled to a single mode of an electromagnetic cavity, are of considerable interest also in connection with quickly developing new quantum physics branches like to cavity quantum electrodynamics [1, 2]. Spin-based solid state quantum bits are known to have long coherence times, while also offering the promise of scalability, and are natural building blocks for quantum computation. Phosphorus donor nuclei in silicon have been known since the 1950s to have some of the best spin coherence properties in solids. The spin coherence time  $T_2$  measured by Hahn spin echo method for donor electron spins in bulk Si:P has been reported to be equal to  $\sim 60$  ms [3]. This is the longest coherence time measured in electron spin qubits and greatly exceeds the values reported, for

instance, the coherence time, observed in GaAs quantum dots, which was measured also by Hahn spin echo method, and it was equal to  $\sim 100 \mu\text{s}$  [4]. However, fabrication of ordered and gated donor arrays and coherent control over donor electrons has turned out to be extremely difficult. There is now much interest in fabricating spin-based devices in diamond, with potential applications in quantum communication, quantum computation, and magnetometry. Nitrogen-vacancy (NV) centers located deep within a diamond lattice appear to be promising solid-state spin qubits since they combine optical initialization and readout capabilities owing to long electron spin coherence life times approaching 1 ms at room temperature [5], and the ability to control coupling to individual nuclear spins. In all of aforesaid applications, it is necessary or at least advantageous to couple NV centers to optical structures like to waveguides and microresonators, to enable communication between distant qubits or to allow efficient extraction of emitted photons. Therefore, a reliable method is needed to create NV centers with good spectral properties in close proximity ( $\leq 100 \text{ nm}$ ) to a diamond surface. In addition, the charge state of the NV center must be controlled. Given tasks are under study at present.

We will show in present work another way to obtain long-lived coherent states with similar field of practical application. The prediction will be based on QED-theory of spectroscopic transitions. The simplest models which capture the salient features of the relevant physics in given field are the Jaynes-Cummings model (JCM) [6] for the one qubit case and its generalization for multiqubit systems by Tavis and Cummings [7]. Tavis-Cummings model was generalized in [8], by taking into account the 1D-coupling between qubits. Recently QED-model for one chain-coupled qubit system was generalized for quasi-one-dimensional axially symmetric multichain-coupled qubit system [9]. It is substantial that in the model, proposed in [9], the interaction of quantized EM-field with multichain qubit system is considered by taking into account both the intrachain and interchain qubit coupling without restriction on their values. It follows from theoretical results in [8–10] and from their experimental confirmation in [11] that by strong interaction of EM-field with matter the correct description of spectroscopic transitions including stationary spectroscopy is achieved only in the frame of QED consideration. It concerns both optical and radio spectroscopies, which means that QED consideration has to be also undertaken by electron spin resonance (ESR) studies in the case of strong interaction of EM-field with spin systems. It is reasonable to suggest that analogous conclusion can be drawn for the case of strong interaction of phonons with spin system or electron system. In other words, there seems to be reasonable the idea that relaxation of optical (or paramagnetic) centers in the case of strong electron-phonon (spin-phonon) coupling can be described correctly only in the frames of quantum field theory.

The aim of the work presented is to derive the system of equations for dynamics of spectroscopic transitions in 1D multiqubit exchange-coupled (para)magnetic and optical systems by strong dipole-photon and dipole-phonon coupling within the framework of quantum electrodynamics

and quantum deformation field theory (phonon theory) and to show that new quantum physics phenomenon—the formation of long-lived coherent state of resonance phonons, leading to appearance of quantum acoustic Rabi oscillations has to be taking place.

## 2. Results and Discussion

Recently in [12], the system of difference-differential equations for dynamics of spectroscopic transitions for both radio and optical spectroscopy for the model, representing itself the 1D-chain of  $N$  two-level equivalent elements coupled by exchange interaction (or its optical analogue for the optical transitions) between themselves and interacting with quantized EM-field and quantized phonon field, has been derived. Naturally the equations are true for any 3D system of paramagnetic centers (PCs) or optical centers by the absence of exchange interaction. In given case the model presented differs from Tavis-Cummings model [7] by inclusion into consideration of quantized phonon system, describing the relaxation processes from quantum field theory position. Seven equations for the seven operator variables, describing joint system {field + matter}, can be presented in matrix form by three matrix equations. They are the following:

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{\sigma}_l^- \\ \hat{\sigma}_l^+ \\ \hat{\sigma}_l^z \end{bmatrix} = 2 \|g\| \begin{bmatrix} \hat{F}_l^- \\ \hat{F}_l^+ \\ \hat{F}_l^z \end{bmatrix} + \|\hat{R}_{ql}^{(\lambda)}\|, \quad (1)$$

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{a}_k^- \\ \hat{a}_k^+ \end{bmatrix} = -i\omega_k \|\sigma_P^z\| \begin{bmatrix} \hat{a}_k^- \\ \hat{a}_k^+ \end{bmatrix} + \frac{i}{\hbar} \begin{bmatrix} -\sum_{l=1}^N (\hat{\sigma}_l^+ + \hat{\sigma}_l^-) v_{lk}^* \\ \sum_{l=1}^N (\hat{\sigma}_l^+ + \hat{\sigma}_l^-) v_{lk}^- \end{bmatrix}, \quad (2)$$

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{b}_q^- \\ \hat{b}_q^+ \end{bmatrix} = -i\omega_q \|\sigma_P^z\| \begin{bmatrix} \hat{b}_q^- \\ \hat{b}_q^+ \end{bmatrix} + \frac{i}{\hbar} \begin{bmatrix} -\sum_{l=1}^N \hat{\sigma}_l^z \lambda_{ql} \\ \sum_{l=1}^N \hat{\sigma}_l^z \lambda_{ql} \end{bmatrix}, \quad (3)$$

where

$$\begin{bmatrix} \hat{\sigma}_l^- \\ \hat{\sigma}_l^+ \\ \hat{\sigma}_l^z \end{bmatrix} = \hat{\sigma}_l = \hat{\sigma}_l^- \vec{e}_+ + \hat{\sigma}_l^+ \vec{e}_- + \hat{\sigma}_l^z \vec{e}_z \quad (4)$$

is vector-operator of spectroscopic transitions for  $l$ th chain unit,  $l = \overline{2, N-1}$  [12]. Its components, that is, the operators

$$\hat{\sigma}_v^{jm} \equiv |j_v\rangle \langle m_v| \quad (5)$$

are set up in correspondence to the states  $|j_v\rangle$ ,  $\langle m_v|$ , where  $v = \overline{1, N}$ ,  $j = \alpha, \beta$ , and  $m = \alpha, \beta$ . For instance, the relationships for commutation rules are

$$[\hat{\sigma}_v^{lm}, \hat{\sigma}_v^{pq}] = \hat{\sigma}_v^{lq} \delta_{mp} - \hat{\sigma}_v^{pm} \delta_{ql}. \quad (6)$$

Further

$$\begin{bmatrix} \hat{F}_l^- \\ \hat{F}_l^+ \\ \hat{F}_l^z \end{bmatrix} = \hat{F} = \left[ \hat{\sigma}_l \otimes \hat{\mathcal{G}}_{l-1,l+1} \right], \quad (7)$$

where vector operators  $\hat{\mathcal{G}}_{l-1,l+1}$ ,  $l = \overline{2, N-1}$ , are given by the expressions

$$\hat{\mathcal{G}}_{l-1,l+1} = \hat{\mathcal{G}}_{l-1,l+1}^- \vec{e}_+ + \hat{\mathcal{G}}_{l-1,l+1}^+ \vec{e}_- + \hat{\mathcal{G}}_{l-1,l+1}^z \vec{e}_z, \quad (8)$$

in which

$$\begin{aligned} \hat{\mathcal{G}}_{l-1,l+1}^- &= -\frac{1}{\hbar} \sum_{\vec{k}} \hat{f}_{l\vec{k}}^- - \frac{J}{\hbar} (\hat{\sigma}_{l+1}^- + \hat{\sigma}_{l-1}^-), \\ \hat{\mathcal{G}}_{l-1,l+1}^+ &= -\frac{1}{\hbar} \sum_{\vec{k}} \hat{f}_{l\vec{k}}^+ - \frac{J}{\hbar} (\hat{\sigma}_{l+1}^+ + \hat{\sigma}_{l-1}^+), \\ \hat{\mathcal{G}}_{l-1,l+1}^z &= -\omega_l - \frac{J}{\hbar} (\hat{\sigma}_{l+1}^z + \hat{\sigma}_{l-1}^z). \end{aligned} \quad (9)$$

Here operator  $\hat{f}_{l\vec{k}}$  is

$$\hat{f}_{l\vec{k}} = v_{l\vec{k}} \hat{a}_{\vec{k}} + \hat{a}_{\vec{k}}^* v_{l\vec{k}}^*. \quad (10)$$

In relations (9)  $J$  is the exchange interaction constant in the case of magnetic resonance transitions or its optical analogue in the case of optical transitions; the function  $v_{l\vec{k}}$  in (10) is

$$v_{l\vec{k}} = -\frac{1}{\hbar} p_l^{jm} (\vec{e}_{\vec{k}}^+ \cdot \vec{e}_{\vec{p}_l}) \mathfrak{E}_{\vec{k}} e^{-i\omega_{\vec{k}} t + i\vec{k}\vec{r}}, \quad (11)$$

where  $p_l^{jm}$  is matrix element of operator of magnetic (electric) dipole moment  $\vec{P}_l$  of  $l$ th chain unit between the states  $|j_l\rangle$  and  $|m_l\rangle$  with  $j \in \{\alpha, \beta\}$ ,  $m \in \{\alpha, \beta\}$ ,  $j \neq m$ ,  $\vec{e}_{\vec{k}}$  is unit polarization vector,  $\vec{e}_{\vec{p}_l}$  is unit vector along  $\vec{P}_l$ -direction,  $\mathfrak{E}_{\vec{k}}$  is the quantity, which has the dimension of magnetic (electric) field strength,  $\vec{k}$  is quantized EM-field wave vector, the components of which get a discrete set of values,  $\omega_{\vec{k}}$  is the frequency, corresponding to  $\vec{k}$ th mode of EM-field, and  $\hat{a}_{\vec{k}}^+$  and  $\hat{a}_{\vec{k}}$  are EM-field creation and annihilation operators correspondingly. In the suggestion that the contribution of spontaneous emission is relatively small, we will have  $p_l^{jm} = p_l^{mj} \equiv p_l$ , where  $j \in \{\alpha, \beta\}$ ,  $m \in \{\alpha, \beta\}$ ,  $j \neq m$ . Further, matrix  $\|\hat{R}_{\vec{q}l}^{(\lambda)}\|$  is

$$\|\hat{R}_{\vec{q}l}^{(\lambda)}\| = \frac{1}{i\hbar} \begin{bmatrix} 2\hat{B}_{\vec{q}l}^{(\lambda)} \hat{\sigma}_l^- \\ -2\hat{B}_{\vec{q}l}^{(\lambda)} \hat{\sigma}_l^+ \\ 0 \end{bmatrix}. \quad (12)$$

Here  $\hat{B}_{\vec{q}l}^{(\lambda)}$  is

$$\hat{B}_{\vec{q}l}^{(\lambda)} = \sum_{\vec{q}} \lambda_{\vec{q}l} (\hat{b}_{\vec{q}}^+ + \hat{b}_{\vec{q}}), \quad (13)$$

$\hat{b}_{\vec{q}}^+$  ( $\hat{b}_{\vec{q}}$ ) is the creation (annihilation) operator of the phonon with impulse  $\vec{q}$  and with energy  $\hbar\omega_{\vec{q}}$ , and  $\lambda_{\vec{q}l}$  is electron-phonon coupling constant. In (2) and (3)  $\|\sigma_{\vec{p}}^z\|$  is Pauli  $z$ -matrix,  $\|g\|$  in (1) is diagonal matrix, and numerical values of its elements are dependent on the basis choice. It is at appropriate basis:

$$\|g\| = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \quad (14)$$

Right-hand side expression in (7) is vector product of vector operators and it is calculated in accordance with the following relation:

$$\left[ \hat{\sigma}_l \otimes \hat{\mathcal{G}}_{l-1,l+1} \right] = \frac{1}{2} \begin{vmatrix} \vec{e}_- \times \vec{e}_z & \hat{\sigma}_l^- & \hat{\mathcal{G}}_{l-1,l+1}^- \\ \vec{e}_z \times \vec{e}_+ & \hat{\sigma}_l^+ & \hat{\mathcal{G}}_{l-1,l+1}^+ \\ \vec{e}_+ \times \vec{e}_- & \hat{\sigma}_l^z & \hat{\mathcal{G}}_{l-1,l+1}^z \end{vmatrix}'. \quad (15)$$

It is seen that vector product of vector operators can be calculated by using known expression for vector product of usual vectors with additional coefficient 1/2 only, which is appeared, since the products of two components of two vectors are replaced in the case of vector operators by anti-commutators of corresponding components. Given detail is mapped by symbol  $\otimes$  in (7) and by symbol  $'$  in determinant in (15).

It follows from comparison with semiclassical Landau-Lifshitz (L-L) equation for dynamics of spectroscopic transitions in a chain of exchange-coupled centers [12, 13] that the equation, which is given by (1), is its QFT-generalization. In comparison with semiclassical description, where the description of dynamics of spectroscopic transitions is exhausted by one vector equation (L-L equation or L-L based equation like (1) in its mathematical form), in the case of completely quantum consideration L-L type equation (1) describes the only one subsystem of three-part system, which consists of EM-field, dipole moments (magnetic or electric) matter subsystem, and phonon subsystem. It was concluded in [12] that the presence of additional equations for description of transition dynamics by QED model in comparison with semiclassical model leads to a number of new effects, which can be predicted only by QED consideration of resonance transition phenomena. One of new effect was described in [12], starting only from the mathematical structure of the equations. It was argued that (1) and (2) represent themselves vector-operator difference-differential generalization of the system, which belongs to well-known family of equation systems—Volterra model systems, widely used in biological tasks of population dynamics studies, which in turn is generalization of Verhulst equation. In other words, it was predicted, for instance, that by some parameters in two-subsystem Volterra model the stochastic component in solution will be appeared. Given prediction has experimental confirmation. So, in [14] the stochastic behavior of joint system {EM-field + matter} was observed by the study of optical properties in carbynes, indicating that in given material strong electron-photon interaction is realized. In other words, quantum description of EM-field

allows to explain the possibility to observe the stationary IR-reflection or absorption spectra both in usual deterministic and in stochastic regime (see for details the next section).

The terms like to right-hand side terms in (3) were used in so-called ‘‘spin-boson’’ Hamiltonian [15] and in so-called ‘‘independent boson model’’ [16]. Given models were used to study phonon effects in a single quantum dot within a microcavity [17–21]. So, the term

$$\widehat{\mathcal{H}}^{\text{CPh}} = \sum_{j=1}^N \sum_{\vec{q}} \lambda_{\vec{q}} (\hat{b}_{\vec{q}}^+ + \hat{b}_{\vec{q}}) \hat{\sigma}_j^z \quad (16)$$

in Hamiltonian in [12] coincides with corresponding term in Hamiltonian in [20, 21] at  $N = 1$ . Contribution of given term to the equations for spectroscopic transitions is  $\pm \sum_{l=1}^N \hat{\sigma}_l^z \lambda_{\vec{q}}$ ; see (3). The equations for spectroscopic transitions were not derived in the previous cited works [17–21]. At the same time, it has been shown in [20, 21] that the presence of given term in Hamiltonian leads to exponential decrease of the magnitude of quantum Rabi oscillations with increase of electron-phonon coupling strength and even to their suppression at relatively strong electron-phonon coupling. However, we will show that by strong electron-photon coupling and strong electron-phonon coupling quite another picture of quantum relaxation processes becomes possible. Really, if to define the wave function of the chain system, interacting with quantized EM-field and with quantized lattice vibration field, to be vector of the state in Hilbert space over quaternion ring, that is, quaternion function of quaternion argument, then like to [12] can be shown that (1) to (3) are Lorentz invariant and the transfer to observables can be realized. In particular, taking into account that quaternion vector of the state is proportional to spin, the Hamiltonian, given by (16), describes in fact (with accuracy to constant factor) the interaction of phonon field with  $z$ -component  $S^z$  of the spin of matter subsystem. It seems to be reasonable to take into consideration the interaction of phonon field with the other peer spin components, that is, with  $S^+$ - and  $S^-$  components of the spin of matter subsystem, which in the case of strong electron-photon coupling seems to be substantial. Therefore, we come in a natural way to the following Hamiltonian:

$$\widehat{\mathcal{H}} = \widehat{\mathcal{H}}^{\text{C}} + \widehat{\mathcal{H}}^{\text{F}} + \widehat{\mathcal{H}}^{\text{CF}} + \widehat{\mathcal{H}}^{\text{Ph}} + \widehat{\mathcal{H}}^{\text{CPh}}, \quad (17)$$

where  $\widehat{\mathcal{H}}^{\text{C}}$  is chain Hamiltonian by the absence of the interaction with EM-field,  $\widehat{\mathcal{H}}^{\text{F}}$  is field Hamiltonian, and  $\widehat{\mathcal{H}}^{\text{CF}}$  is Hamiltonian, describing the interaction between quantized EM-field and atomic chain. Hamiltonian  $\widehat{\mathcal{H}}^{\text{C}}$  is

$$\widehat{\mathcal{H}}^{\text{C}} = \widehat{\mathcal{H}}^0 + \widehat{\mathcal{H}}^{\text{J}}, \quad (18)$$

where  $\widehat{\mathcal{H}}^0$  is chain Hamiltonian in the absence of the interaction between structural elementary units of the chain.  $\widehat{\mathcal{H}}^0$  is given by the following expression:

$$\widehat{\mathcal{H}}^0 = \sum_{v=1}^N \sum_m E_{mv} |m_v\rangle \langle m_v|. \quad (19)$$

Here  $m = \alpha, \beta$ ,  $E_{mv}$  are eigenvalues of  $\widehat{\mathcal{H}}^0$ , which correspond to the states  $|m_v\rangle$  of  $v$ th chain unit. Hamiltonian  $\widehat{\mathcal{H}}^{\text{J}}$  is

$$\widehat{\mathcal{H}}^{\text{J}} = \sum_{n=1}^N \left[ J_E \left( \hat{\sigma}_n^+ \hat{\sigma}_{n+1}^- + \hat{\sigma}_n^- \hat{\sigma}_{n+1}^+ + \frac{1}{2} \hat{\sigma}_n^z \hat{\sigma}_{n+1}^z \right) + \text{H.c.} \right]. \quad (20)$$

It is suggested in the model that  $|\alpha_n\rangle$  and  $|\beta_n\rangle$  are eigenstates, producing the full set for each of  $N$  elements. It is evident that given assumption can be realized strictly only by the absence of the interaction between the elements. At the same time proposed model will rather well describe the real case, if the interaction energy of adjacent elements is much less of the energy of the splitting  $\hbar\omega_0 = \mathcal{E}_\beta - \mathcal{E}_\alpha$  between the energy levels, corresponding to the states  $|\alpha_n\rangle$  and  $|\beta_n\rangle$ . The case considered includes in fact all known experimental situations. It is clear that Hamiltonian  $\widehat{\mathcal{H}}^{\text{CF}}$  of interaction of quantized EM-field with atomic chains can also be represented in the set of variables, which includes the components of spectroscopic transition vector operator  $\hat{\sigma}_v$ . Really, suggesting dipole approximation to be true and polarization of field components to be fixed, we have

$$\widehat{\mathcal{H}}^{\text{CF}} = - \sum_{j=1}^n \sum_{l \neq m} \sum_m \sum_{\vec{k}} \left[ p_j^{lm} \hat{\sigma}_j^{lm} \left( \vec{e}_{\vec{k}} \cdot \vec{e}_{\vec{p}_j} \right) \mathfrak{E}_{\vec{k}} \hat{a}_{\vec{k}} \times e^{-i\omega_{\vec{k}}t + i\vec{k}\vec{r}} + \text{H.c.} \right], \quad (21)$$

where  $p_j^{lm}$  is matrix element of operator of magnetic (electric) dipole moment  $\vec{P}_j$  of  $j$ th chain unit between the states  $|l_j\rangle$  and  $|m_j\rangle$  with  $l_j = \alpha_j, \beta_j$ ,  $m_j = \alpha_j, \beta_j$ ,  $\vec{e}_{\vec{k}}$  is unit polarization vector,  $\vec{e}_{\vec{p}_j}$  is unit vector along  $\vec{P}_j$ -direction,  $\mathfrak{E}_{\vec{k}}$  is the quantity, which has the dimension of magnetic (electric) field strength,  $\vec{k}$  is wave vector, and  $\hat{a}_{\vec{k}}$  is field annihilation operator. In the suggestion that the contribution of spontaneous emission is relatively small, we will have  $p_j^{lm} = p_j^{ml} \equiv p_j$ , where  $l = \alpha, \beta$ , and  $m = \alpha, \beta$ . Let us define the function

$$q_{j\vec{k}} = - \frac{1}{\hbar} p_j \left( \vec{e}_{\vec{k}} \cdot \vec{e}_{\vec{p}_j} \right) \mathfrak{E}_{\vec{k}} e^{-i\omega_{\vec{k}}t + i\vec{k}\vec{r}}. \quad (22)$$

Then the expression (21) can be rewritten in the following form:

$$\widehat{\mathcal{H}}^{\text{CF}} = \sum_{v=1}^n \sum_{\vec{k}} \left[ q_{j\vec{k}} (\hat{\sigma}_j^- + \hat{\sigma}_j^+) \hat{a}_{\vec{k}} + (\hat{\sigma}_j^- + \hat{\sigma}_j^+) \hat{a}_{\vec{k}}^* q_{j\vec{k}}^* \right], \quad (23)$$

where  $\hat{a}_{\vec{k}}^+$  is EM-field creation operator, and superscript  $*$  in  $q_{j\vec{k}}^*$  means complex conjugation. Field Hamiltonians have usual form

$$\widehat{\mathcal{H}}^{\text{F}} = \sum_{\vec{k}} \hbar\omega_{\vec{k}} \left( \hat{a}_{\vec{k}}^+ \hat{a}_{\vec{k}} + \frac{1}{2} \right) \quad (24)$$

for EM-field and

$$\widehat{\mathcal{H}}^{\text{Ph}} = \sum_{\vec{q}} \hbar\omega_{\vec{q}} \left( \hat{b}_{\vec{q}}^+ \hat{b}_{\vec{q}} + \frac{1}{2} \right) \quad (25)$$

for phonon field, where  $\hat{b}_q^+$  and  $\hat{b}_q^-$  are phonon-field creation and annihilation operators, respectively. Hamiltonian  $\hat{\mathcal{H}}^{\text{CPh}}$  is

$$\hat{\mathcal{H}}^{\text{CPh}} = \hat{\mathcal{H}}_z^{\text{CPh}} + \hat{\mathcal{H}}_{\pm}^{\text{CPh}}, \quad (26)$$

where  $\hat{\mathcal{H}}_z^{\text{CPh}}$  is determined by the following expression:

$$\hat{\mathcal{H}}_z^{\text{CPh}} = \sum_{j=1}^N \sum_{\bar{q}} \left( \lambda_{\bar{q}}^z \hat{b}_{\bar{q}}^+ + (\lambda_{\bar{q}}^z)^* \hat{b}_{\bar{q}}^- \right) \hat{\sigma}_j^z. \quad (27)$$

Hamiltonian  $\hat{\mathcal{H}}_{\pm}^{\text{CPh}}$  can be represented in the following form:

$$\hat{\mathcal{H}}_{\pm}^{\text{CPh}} = \sum_{j=1}^N \sum_{\bar{q}} \lambda_{\bar{q}}^{\pm} (\hat{\sigma}_j^- + \hat{\sigma}_j^+) \hat{b}_{\bar{q}}^{\pm} + (\lambda_{\bar{q}}^{\pm})^* (\hat{\sigma}_j^- + \hat{\sigma}_j^+) \hat{b}_{\bar{q}}^{\pm}. \quad (28)$$

Here  $\lambda_{\bar{q}}^z$  and  $\lambda_{\bar{q}}^{\pm}$  are electron-phonon coupling constants, which characterise correspondingly the interaction with  $z$ -component  $S_j^z$  and with  $S_j^+$ - and  $S_j^-$  components of the spin of  $j$ th chain unit. It seems to be understandable that they can be different in general case. Moreover, in order to take into account the interaction with both equilibrium and nonequilibrium phonons, both the electron-phonon coupling constants have to be complex numbers that takes proper account by expressions (27) and (28).

It can be shown that the equations of the motion for spectroscopic transition operators  $\hat{\sigma}_l$ , for quantized EM-field operators  $\hat{a}_k^+$ ,  $\hat{a}_k^-$  and for phonon field operators  $\hat{b}_q^+$ ,  $\hat{b}_q^-$  are the following. Instead of (1), we have

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{\sigma}_l^- \\ \hat{\sigma}_l^+ \\ \hat{\sigma}_l^z \end{bmatrix} = 2 \|g\| \begin{bmatrix} \hat{F}_l^- \\ \hat{F}_l^+ \\ \hat{F}_l^z \end{bmatrix} + \|\hat{R}_{\bar{q}l}^{(\lambda^z)}\| + \|\hat{R}_{\bar{q}l}^{(\lambda^{\pm})}\|, \quad (29)$$

where matrix  $\|\hat{R}_{\bar{q}l}^{(\lambda^z)}\|$  is

$$\|\hat{R}_{\bar{q}l}^{(\lambda^z)}\| = \frac{1}{i\hbar} \begin{bmatrix} 2\hat{B}_{\bar{q}l}^{(\lambda^z)} \hat{\sigma}_l^- \\ -2\hat{B}_{\bar{q}l}^{(\lambda^z)} \hat{\sigma}_l^+ \\ 0 \end{bmatrix} \quad (30)$$

with  $\hat{B}_{\bar{q}l}^{(\lambda^z)}$ , which is given by

$$\hat{B}_{\bar{q}l}^{(\lambda^z)} = \sum_{\bar{q}} \left[ (\lambda_{\bar{q}l}^z)^* \hat{b}_{\bar{q}}^+ + \lambda_{\bar{q}l}^z \hat{b}_{\bar{q}}^- \right]. \quad (31)$$

Matrix  $\|\hat{R}_{\bar{q}l}^{(\lambda^{\pm})}\|$  is

$$\|\hat{R}_{\bar{q}l}^{(\lambda^{\pm})}\| = \frac{1}{i\hbar} \begin{bmatrix} -\hat{B}_{\bar{q}l}^{(\lambda^{\pm})} \hat{\sigma}_l^z \\ \hat{B}_{\bar{q}l}^{(\lambda^{\pm})} \hat{\sigma}_l^z \\ \hat{B}_{\bar{q}l}^{(\lambda^{\pm})} (\hat{\sigma}_l^+ - \hat{\sigma}_l^-) \end{bmatrix}, \quad (32)$$

where  $\hat{B}_{\bar{q}l}^{(\lambda^{\pm})}$  is

$$\hat{B}_{\bar{q}l}^{(\lambda^{\pm})} = \sum_{\bar{q}} \left[ (\lambda_{\bar{q}l}^{\pm})^* \hat{b}_{\bar{q}}^+ + \lambda_{\bar{q}l}^{\pm} \hat{b}_{\bar{q}}^- \right]. \quad (33)$$

Equation (2) remains without changes. Equation (3) is

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{b}_{\bar{q}}^- \\ \hat{b}_{\bar{q}}^+ \end{bmatrix} = -i\omega_{\bar{q}} \|\sigma_{\bar{p}}^z\| \begin{bmatrix} \hat{b}_{\bar{q}}^- \\ \hat{b}_{\bar{q}}^+ \end{bmatrix} + \frac{i}{\hbar} \begin{bmatrix} -\sum_{l=1}^N \left\{ \lambda_{\bar{q}l}^z \hat{\sigma}_l^z + \lambda_{\bar{q}l}^{\pm} (\hat{\sigma}_l^+ + \hat{\sigma}_l^-) \right\} \\ \sum_{l=1}^N \left\{ \lambda_{\bar{q}l}^z \hat{\sigma}_l^z + \lambda_{\bar{q}l}^{\pm} (\hat{\sigma}_l^+ - \hat{\sigma}_l^-) \right\} \end{bmatrix}. \quad (34)$$

Thus, QFT model for dynamics of spectroscopic transitions in 1D multiqubit exchange-coupled system is generalized by taking into account the earlier proof [12] that spin vector is quaternion vector of the state of any quantum system in Hilbert space defined over quaternion ring and consequently all the spin components have to be taken into account. New quantum phenomenon is predicted. The prediction results from the structure of the equations derived and it consists in the following. The coherent system of the resonance phonons, that is, the phonons with the energy, equaled to resonance photon energy can be formed by resonance that can lead to appearance along with Rabi oscillations determined by spin- (electron-) photon coupling with the frequency  $\Omega^{\text{RF}}$  of Rabi oscillations determined by spin- (electron-) phonon coupling with the frequency  $\Omega^{\text{RPh}}$ . In other words QFT model predicts the oscillation character of quantum relaxation that is quite different character in comparison with phenomenological and semiclassical Bloch models. Moreover if  $|\lambda_{\bar{q}l}^{\pm}| < q_{\bar{q}l}$ , the second Rabi oscillation process will be observed by stationary state of two subsystems {EM-field + magnetic (electric) dipoles}; that is, it will be registered in quadrature with the first Rabi oscillation process. It can be experimentally detected even by stationary spectroscopy methods.

In the case of strong electron-photon and strong electron-phonon couplings, the joint {EM-field + magnetic dipole + resonance phonon field} system has to be considered. EM-field and resonance phonon field produce radiation communication in the direction of EM-field propagation and in time. It means that we have 1D chain, at that the chains of two kinds are produced in time—with the sequence absorption time segment and emission time segment and with reversal sequence. It is substantially that given time segments are different in their values. In fact, the dimerisation takes place like to transpolyacetylene, however instead space  $z$ -axis along  $t$ -axis. It is in agreement with known equality of rights of coordinates in Minkowski space. In other words, the time has to be considered being to be quantized (dimerized) in given case. The junction of given time ordered chains produces topological time violations of two kinds. The first topological violation can effectively absorb photons and emit resonance phonons, and the second

topological violation has reversal properties. So, we come to the model of instantons, which is like to some extent to SSH-soliton model. It is understandable that the instantons of both kinds can free, that is, without additional activation energy, move along  $t$ -axis like SSH-solitons along space  $z$ -axis. Given model allows to explain the rather narrow magnetic resonance or optical lines in structurally ordered or even disordered materials in the following way. The instantons seem to be two-level objects, which are noninteracting between themselves and with surrounding (dipole subsystem of instantons in excited state interacts with surrounding very short time, that determines minimal role of fluctuations in surrounding). Consequently, instantons will be characterised by discrete energetic structure with very narrow energy dispersion, which determines in turn the small resonance line broadening. In other words, the instanton model explains the appearance of narrow resonance lines in some condensed matter systems in a natural way. So QFT-model allows to predict the instantons of new kind and the appearance of very narrow lines in some resonance spectra.

The known instantons are considered in the literature to be a special kind of vacuum oscillations in gluon field theory and also in gravitation field theory [22]. They have quite another origin.

### 3. Comparison with Experiment

More detailed description of experimental results, partly reported earlier in [11, 12, 14], will be now given. The matter concerns the results, which can be explained only by taking into account the quantum nature of EM-field.

To compare the QFT-model of spectroscopic transitions, referred to Volterra model systems, with experiment the parameters of the model, in particular the ratios between  $\lambda_{ni}^{\pm}$ ,  $\lambda_{ni}^z$  and  $q_{ni}$ , have to be varied during the experiment. It can be achieved by the change of velocity of passage through the resonance, taking into account the topological nature of optical and magnetic resonance centers in the samples studied (see for details further). The change of velocity of passage through the resonance was achieved by the change of the access velocity by digital treatment of optical signals. Infrared (IR) absorption and reflection studies have been performed on uniaxially oriented carbynoid film samples prepared by chemical dehydrohalogenation of poly(vinylidene fluoride) (PVDF) films, which were stored 12 Y at room temperature. Preparation details of carbynoids are described in [23]. The FTIR spectrometer “Nexus” has been used, and IR spectra were registered in the range 400–5000  $\text{cm}^{-1}$  at room temperature. Two groups of uniaxially oriented carbynoid film samples were studied, designated correspondingly A and B samples (the second and the third series samples according to designation in [23]). They were differed by the contamination of fluorine and oxygen atoms; see for details [23]. Like to the classification of organic conductors proposed for doped *trans*-polyacetylene (*t*-PA) [24], the samples studied can be attributed both to highly doped carbynes and to carbynoids that is to materials including a wide range of carbyne-like structures.

It was established that the difference of the spectra for the samples, belonging to the same group, is in limits of uncertainty of experimental measurements. Moreover, the dependence of the spectra on the sweep velocity through the resonance was qualitatively similar for the two groups, and in given section, the results only for the B-sample will be represented.

It has been found earlier that topological polarons in C–C  $\sigma$ -bonds of organic polymers can be formed. In particular, in carbynoids and in polyvinylidene fluoride, they were found to be electric own dipole (called electric spin moment) carriers and are considered to be representative of new  $\sigma$ -quasiparticles’ family [14]. Let us remark that given family allows to supplement the well-known Su-Schrieffer-Heeger (SSH) model of organic polymers, which describes the formation of topological  $\pi$ -quasiparticles [25] to be spin or charge carriers.

There has been reported on the observation of IR reflection in uniaxially oriented carbynoid films for the first time in [26]. Detailed analysis, performed in the range (800–2200)  $\text{cm}^{-1}$ , has shown that two sets of the components in given range represent themselves the spectra, displaying new physics phenomenon—ferroelectrical spin wave resonance (FESWR). Let us remark the following. It was established earlier [23, 27] that the main charge and spin carriers in carbynoids are topological  $\pi$ -solitons called spin-Peierls solitons (SPSs). SPSs possess like topological solitons in *t*-PA the vibration activity. IR-lines, which were attributed, respectively, to localized IR-active SPS vibration modes  $a$ ,  $b$ , and  $c$  in particular in B-samples, have the positions at 1750, 1650, and 1080  $\text{cm}^{-1}$  [14, 28–30]. Both the SPS high-frequency vibration modes  $a$  and  $b$  split into sets  $\{a_n\}$  and  $\{b_n\}$ . The components with  $n = \overline{1,3}$  were identified. For instance, in B-sample the sets  $\{a_n\}_{[A]}$  and  $\{b_n\}_{[A]}$  are observed in result of splitting of  $a$ - and  $b$ -lines, and they, correspondingly, are  $\{a_n\}_{[B]} = \{1738.3, 1464.0, 903.6 \text{ cm}^{-1}\}$ , and  $\{b_n\}_{[B]} = \{1685.7, 1419.4, 886.7 \text{ cm}^{-1}\}$  (Figure 1). The third SPS-mode ( $c$ -line) is observed at 1088.4  $\text{cm}^{-1}$ . It is however weakly pronounced and its splitting into set like to splitting of  $a$ - and  $b$ -lines was not detected.

It was found that just the splitting observed is the most substantial characteristic of given new quantum optics phenomenon, that is, FESWR. The values of FESWR splitting parameter  $\mathfrak{A}$  in dispersion law amount  $\mathfrak{A}_{a[B]} = 34.7 \text{ cm}^{-1}$  and  $\mathfrak{A}_{b[B]} = 33.2 \text{ cm}^{-1}$  for the sample of B-set. Observation of FESWR on SPS vibration modes means that electric dipole moments of SPS in carbynoid chains are ferroelectrically ordered in result of soliton-soliton interaction. It seems to be very interesting that carbynes along with ferroelectric ordering possess also by antiferroelectric and ferromagnetic ordering. The conclusion on ferromagnetic ordering has been obtained from ESR studies of heavily doped by technological impurities carbynes in [23, 27] by means of immediate observation of ferromagnetic spin wave resonance. The conclusion on antiferroelectric ordering of carbynes was also obtained and also by direct registration in given case of antiferroelectric spin wave resonance (AFESWR), that was reported in [14, 30], especially interesting, that antiferroelectricity, ferroelectricity, and ferromagnetism have been

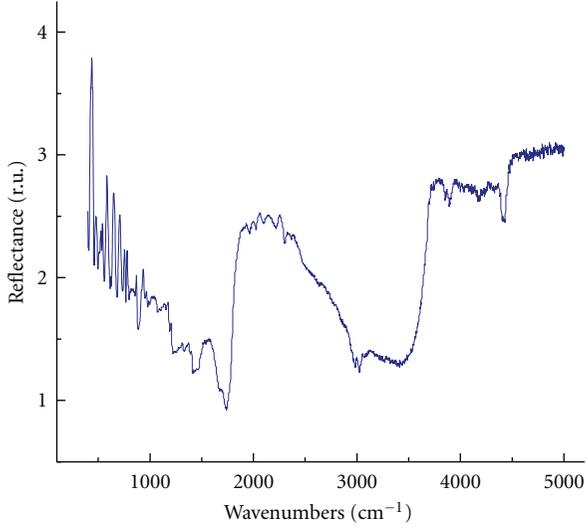


FIGURE 1: Spectral distribution of IR reflection intensity in carbynoid sample in the range 400–5000  $\text{cm}^{-1}$ , registered in deterministic regime.

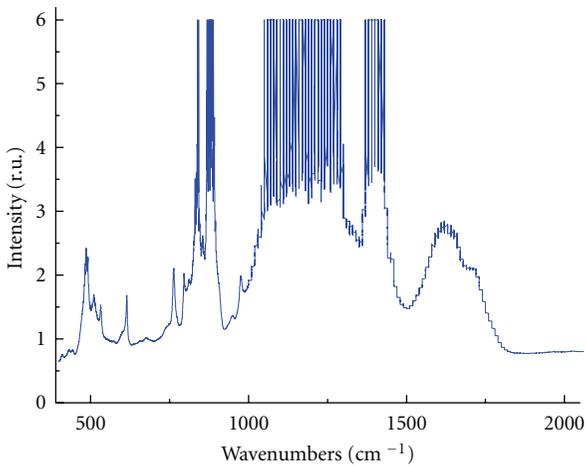


FIGURE 2: Spectral distribution of IR absorption intensity in carbynoid sample in the range 400–2100  $\text{cm}^{-1}$ , registered in intermediate regime (between deterministic and stochastic ones).

observed simultaneously in the same samples and in the same room temperature range.

Topological  $\sigma$ -polaron lattice was proposed to be responsible for the observed AFESWR both in starting PVDF films and in carbynoid films. It is reasonable to suggest that dynamical systems of  $\sigma$ -polarons, which are responsible for AFESWR-spectrum in the range 400– $\approx$ 900  $\text{cm}^{-1}$ , and  $\pi$ -solitons, which are responsible for the ferroelectric spin wave resonance [26], in particular, for the spectrum in the range 1500–1870  $\text{cm}^{-1}$ , Figure 1, and for broad absorption line in the range 1800–3700  $\text{cm}^{-1}$ , Figure 1 [28, 31], are strongly coupled with photons by the interaction with external EM-field. It means that correct description of optical transitions will be achieved only in the frame of QFT-model proposed. Moreover, in given case the number of different experimental

registration regimes can be realized. In particular, optical spectra can be registered in stochastic regime. In favour of given conclusions is indicated the appearance of the lines in IR spectra in the range (3500–5000)  $\text{cm}^{-1}$ . Their peak positions are the following—3892.6  $\pm$  4  $\text{cm}^{-1}$ , 4174.6  $\pm$  4  $\text{cm}^{-1}$ , and 4420.3  $\pm$  4  $\text{cm}^{-1}$  in the spectrum of B-sample. The appearance of lines in high-frequency spectral range is usually attributed to so-called second-order transitions. We see, however, that any combination of the frequency values for the lines in low-frequency range (800–2200)  $\text{cm}^{-1}$ , which potentially can be considered to be the range of the first order transitions, cannot explain the foregoing values of the frequencies of the lines, observed in the range (3500–5000)  $\text{cm}^{-1}$ . It was shown in [11] that the appearance of lines in high-frequency spectral range (3500–5000)  $\text{cm}^{-1}$  in carbynoid samples is the result of quantum nature of EM-field leading to emergence of new coherent quantum optics phenomenon—Rabi wave packets' formation and propagation. Given lines have to be disappeared by the registration in stochastic regime to be consequence of coherence breakdown. The results, represented in Figures 3, 4, and 6 indicate that it really takes place. Let us comment on given results (along with the results represented in Figures 2 and 5 in more details). It was shown in [26] by means of comparison of the absorption (transmission) and reflection spectra in the range, where the absorption is not very strong, that reflection in the samples studied has bulk character and the reflection spectra after signal polarity change and amplitude renormalization by appropriate scale factor become practically coinciding with the absorption (transmission) spectra; compare, for instance, Figure 1 in given paper and Figure 1 in [12] in the range 3000–5000  $\text{cm}^{-1}$ . It is seen from given figures that the lines at 3892.6  $\pm$  4  $\text{cm}^{-1}$ , 4174.6  $\pm$  4  $\text{cm}^{-1}$ , and 4420.3  $\pm$  4  $\text{cm}^{-1}$  are presenting in both transmission and reflection spectra by the usual registration conditions in deterministic regime. At the same time, they are disappeared by the increasing of sweep velocity by a factor of 42 and the transfer to stochastic regime; see Figure 3. The spectral distribution of IR absorption intensity in carbynoid sample in the range 400–2100  $\text{cm}^{-1}$ , registered in intermediate between deterministic and stochastic regime (sweep velocity increase is characterised by a factor of 4.2), is represented in Figure 2. It is seen that the lines in the ranges 400–830 and 900–1000  $\text{cm}^{-1}$  are registered in deterministic regime (they correspond to the modes of AFESWR resonance, determined by  $\sigma$ -subsystem of sample vibronic system). At the same time, the lines in the ranges 830–900 and 1000–5000  $\text{cm}^{-1}$  are registered already in stochastic regime, which is characterized by the appearance of specific quantum noises, at that the amplitude of the lines at 3892.6  $\pm$  4  $\text{cm}^{-1}$ , 4174.6  $\pm$  4  $\text{cm}^{-1}$ , and 4420.3  $\pm$  4  $\text{cm}^{-1}$  is strongly decreasing in intermediate regime (it is not shown in Figure 2). It was found that the amplitude of quantum noises is strongly dependent on wave number; see Figures 4 to 6. It is interesting, that spectral distribution of noises corresponds to counters of the lines, which are surviving in stochastic regime. Moreover, even the shape of the nonoverlapped lines can be determined from amplitude dependence of quantum noises on wave number.

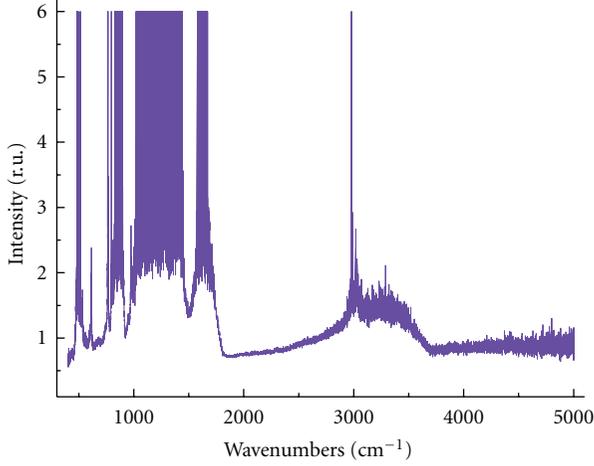


FIGURE 3: Spectral distribution of IR absorption intensity in carbynoid sample in the range 400–5000  $\text{cm}^{-1}$ , registered in stochastic regime.

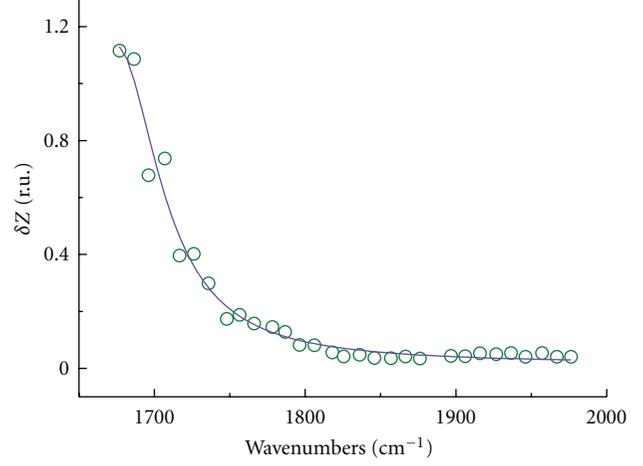


FIGURE 5: Lorentz shape fit of spectral distribution of “noise”-amplitude of IR absorption in carbynoid sample in the range 1675–1870  $\text{cm}^{-1}$ , corresponding to right wing of the main  $a$ -mode in FESWR on spin-Peierls- $\pi$ -solitons.

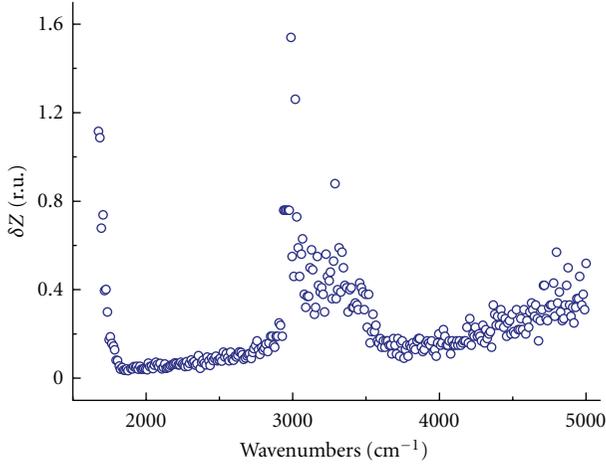


FIGURE 4: Spectral distribution of “noise”-amplitude of IR absorption intensity in carbynoid sample in the range 1675–5000  $\text{cm}^{-1}$ , registered in stochastic regime.

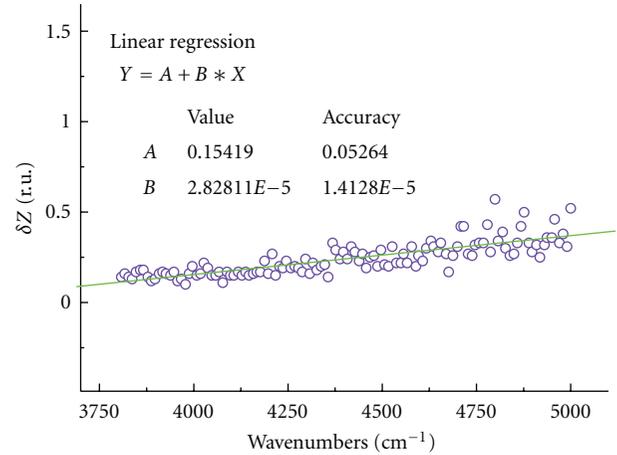


FIGURE 6: Spectral distribution of “noise”-amplitude of IR absorption intensity in carbynoid sample in the range 3750–5000  $\text{cm}^{-1}$ , registered in stochastic regime.

For instance, in Figure 5, the fit of spectral distribution of “noise”-amplitude of IR absorption in carbynoid sample in the range 1675–1870  $\text{cm}^{-1}$ , corresponding to right wing of the main FESWR- $a$ -mode of SPS- $\pi$ -solitons, by Lorentz shape is represented. It is seen, Figure 3, that the lines at  $3892.6 \pm 4 \text{ cm}^{-1}$ ,  $4174.6 \pm 4 \text{ cm}^{-1}$ , and  $4420.3 \pm 4 \text{ cm}^{-1}$  are disappeared entirely in stochastic regime. They cannot be extracted even from analysis of spectral distribution of quantum noises; see Figure 6. It is seen from given figure, that the only quantum noises are observed in the range 3750–5000  $\text{cm}^{-1}$ , and their spectral distribution in given range can be approximated by linear dependence.

Therefore, the experimental results previously described are strong indication on necessity to take into consideration the quantum nature of EM-field by the study of dynamics of spectroscopic transitions, and they can be considered to

be experimental confirmation of mathematical structure of equations in QFT-model proposed.

## 4. Conclusions

QFT-model for dynamics of spectroscopic transitions in 1D multiqubit exchange-coupled system is generalized by taking into account the earlier proof [12] that spin vector is quaternion vector of the state of any quantum system in Hilbert space defined over quaternion ring, and consequently all the spin components have to be taken into account. New quantum phenomenon is predicted. The prediction results from the structure of the equations derived and it consists in the following. The coherent system of the resonance phonons, that is, the phonons with the energy, equaled to resonance photon energy can be formed by resonance

that can lead to appearance along with Rabi oscillations determined by spin- (electron-) photon coupling with the frequency  $\Omega^{RF}$  of Rabi oscillations determined by spin- (electron-) phonon coupling with the frequency  $\Omega^{RPh}$ . In other words, QFT model predicts the oscillation character of quantum relaxation that is quite different character in comparison with phenomenological and semiclassical Bloch models. Moreover if absolute value of electron-phonon coupling constant  $|\lambda_{qj}^{\pm}|$ , which characterises the interaction with  $S^+$ - and  $S_j^-$  components of the spin of  $j$ th chain unit, is less than electron- (spin-) photon coupling constant, the model predicts that the second quantum Rabi oscillation process will be observed by stationary state of joint two subsystems {EM-field + magnetic (electric) dipoles}, and it will be registered in quadrature with the first quantum Rabi oscillation process. The second quantum Rabi oscillation process is governed by the formation of the coherent system of the resonance phonons. Therefore along with absorption process of EM-field energy, the coherent emission process takes place. Both the quantum Rabi oscillation processes can be time shared. For the case of radio spectroscopy, it corresponds to the possibility of the simultaneous observation along with (para)magnetic spin resonance the acoustic spin resonance. The second (acoustic) quantum Rabi oscillation process can be detected even by stationary spectroscopy methods.

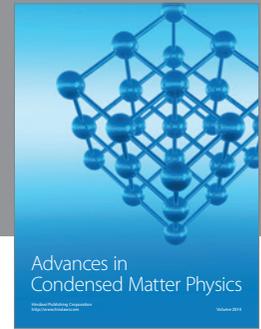
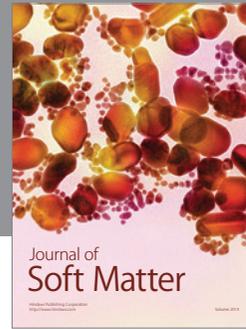
The predicted phenomenon of the formation of the coherent system of the resonance phonons can find the number of practical applications; in particular it can be used by elaboration of various logic quantum systems including quantum computers and quantum communication systems, in which quantized EM-field and/or quantized acoustic field will be working components. The same conclusion is concerned of the elaboration of various optoelectronic and spintronic devices.

The comparison with some experimental results is represented.

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