On the vibron-polaron damping in quasi 1D macromolecular chains

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Abstract. The properties of the intramolecular vibrational excitation (vibron) in a quasi 1D macromolecular structure are studied. It is supposed that due to the vibron interaction with optical phonon modes, a vibron might form partially dressed small polaron states. The properties of these states are investigated in dependence on the basic system parameters and temperature of a thermal bath. We also investigate the process of damping of the polaron amplitude as a function of temperature and vibron-phonon coupling strength. Two different regimes of the polaron damping are found and discussed.

1. Introduction
The knowledge of exciton properties and especially physics of exciton transport in quasi 1D structures is very important in many fields of material sciences. Here, it is pertinent to mention the problem of charge and energy transport in biological macromolecules (MCs) and the problem of the quantum information transport in quantum circuits, especially in realistic conditions, where a thermal bath is an important source of quantum information losses. For example, the energy and charge transport in biological macromolecules (such as protein macromolecules, RNA, and DNA) plays an important role in the processes which take place in functioning of living cells. One of the most important processes is the photosynthesis, in which a photon quantum is absorbed by special cell structures (light-harvesting antennas) as an electronic excitation. After that these excitations are transported to the reaction centres, where they are converted to the chemical energy. It is remarkable fact that during its migration almost every absorbed photon is successfully transferred to the reaction centre despite its short life-time (the characteristic life-time of an electron excitation is approximately 1 ns).

There are two main subsystems that we are interested in, namely a vibronic excitation and phonons. Due to the presence of intersite coupling, a vibronic excitation can migrate between two adjacent sites. The phonon subsystem affects to the excitation migration on different ways. First, due to the presence of the phonon-excitation interaction, the excitation can be self-trapped and form a new quasiparticle dressed by the cloud of virtual phonons, i.e. a polaron quasiparticle. Under some circumstances, such a quasiparticle can be more stable than the bare excitation, and
in principle such a model might explain the transfer of the excitation over long distances along a macromolecular spine [1, 2, 3, 4]. Second, the phonon subsystem is a source of dissipative processes in relation to the excitation (polaron) subsystem. Phonons prevent the formation of the polaron quasiparticle and reduce its life-time. As a consequence, the transfer properties of polarons in MCs are determined by the interplay of dynamic and dissipative processes [3, 5, 6].

In this paper, we investigate the process of relaxation of exciton states on the lattice vibrations in 1D macromolecular chains. We suppose that, due to the presence of exciton-phonon coupling, an exciton quasiparticle is self-trapped and forms a non-adiabatic polaron state. It is then assumed that the dissipation of such a state is the consequence of the presence of the residual polaron-phonon interaction (all other sources of dissipation, such as polaron-polaron interaction, are disregarded). The process of the exciton self-trapping is described in the Sec. 2. Main properties of the formed polaron state are presented in dependence on the values of the basic energy parameters of macromolecular systems. In the Sec. 3 we consider the dependence of the polaron amplitude damping on the system parameters and the thermal bath temperature.

2. Physical foundation of the problem
The framework of our investigation is the Holstein molecular crystal model (MCM) [7]. We start from the Hamiltonian which includes the exciton (vibron) interaction with the optical phonon subsystem.

\[
H = E \sum_n a_n^\dagger a_n - J \sum_n a_n^\dagger (a_{n-1} + a_{n+1}) + \sum_q \hbar \omega_q b_q^\dagger b_q + \frac{1}{\sqrt{N}} \sum_{n,q} F_q e^{iq R_0} a_n^\dagger a_n \left( b_q + b_{-q}^\dagger \right). \tag{1}
\]

In the above Hamiltonian \(E\) is the exciton excitation energy, \(J\) is the resonant transfer integral (intersite coupling strength), which is responsible for quantum jumps of the particle between neighbouring sites, \(R_0\) is the parameter of the macromolecular chain, \(a_n\) (\(a_n^\dagger\)) are the operators of annihilation (creation) of an exciton on the \(n\)-th structure element of the macromolecular chain, and \(b_q\) (\(b_q^\dagger\)) are the phonon annihilation (creation) operators. The relation for the exciton-phonon coupling constant \(F_q\) depends on the nature of the phonon subsystem. For the optical phonons, this parameter has the form \(F_q = \chi \sqrt{\hbar/(2M \omega_q)}\) with \(\chi\) and \(M\) being by the interaction parameter and the mass of the chain structure element respectively.

The standard procedure to pass into the polaron picture (dressed vibron point of view), is based on the Lang-Firsov (LF) unitary transformation [4, 8]. Such approach is applicable in the case where the conditions for a formation of the nonadiabatic polaron are fulfilled (i.e. for the strong-coupling exciton–phonon case). However, in the case of a vibron excitation in the biological macromolecules these conditions are not satisfied (and therefore vibron-phonon interaction can belong to intermediate or weak coupling cases!) [9]. Consequently, in this case, the variational approaches are much more promising ones [10, 11, 12, 13, 14]. Here, we use the variational approach based on the modified Lang-Firsov unitary transformation (MLFUT): \(U = \exp(-\sum_n a_n^\dagger a_n S_n)\), where \(S_n = 1/\sqrt{N} \sum_q f_q e^{-iq R_0} (b_q - b_{-q}^\dagger)\), and \(f_q = f_{x_q}\) being by the parameters that determine the degree of exciton dressing by a cloud of phonons. They can be obtained with use of the variational approach, i.e. by the minimization of the polaron ground state energy. Using MLFUT, we obtain a transformed Hamiltonian with new operators that correspond to dressed exciton (polaron) operators and the new phonons (which describe oscillations of structure elements around new equilibrium positions):

\[
\tilde{H} = \mathcal{E} \sum_n a_n^\dagger a_n - J \sum_n a_n^\dagger (a_{n-1} \theta_{n-1} + a_{n+1} \theta_{n+1}) + \sum_n \hbar \omega_q b_q^\dagger b_q + \\
+ \frac{1}{\sqrt{N}} \sum_{n,q} \left( F_q - \hbar \omega_q f_{x_q}^* \right) e^{iq R_0} a_n^\dagger a_n \left( b_q + b_{-q}^\dagger \right) + \tilde{H}_R, \tag{2}
\]
where $\theta_{n+1} = \exp(S_{n+1} - S_n)$ is dressing operator, and $\mathcal{E} = E + N^{-1} \sum q \hbar \omega_q |f_q|^2 - N^{-1} \sum q F_q(f_q + f^*_q)$ is the energy shift of dressed excitons. The fourth term in (2) describes the remaining interaction between the polaron and new phonon, which is responsible for the phonon-induced polaron dissipation processes (i.e. for polaron amplitude damping). The rest part of the Hamiltonian reads as $\hat{H}_R = N^{-1} \sum_{q,n,m} \left\{ \hbar \omega_q |f_q|^2 - F_q(f_q + f^*_q) \right\} e^{i q (n-m) R_0} a_n a_m^\dagger$ and corresponds to the multi-polaron processes.

In order to account for the influence of thermal fluctuations of structure elements on polaron properties, we make averaging of our Hamiltonian over the new phonon subsystem to get $\hat{H} = \hat{H}_0 + \hat{H}_{\text{rest}}$. Here, $\hat{H}_0 = \hat{H}_{\text{vib}} + \hat{H}_{\text{ph}}$ stands for the effective mean-field Hamiltonian consists of effective vibron ($\hat{H}_{\text{vib}} = \left\langle \hat{H} - \hat{H}_{\text{ph}} \right\rangle_{\text{ph}}$) and the phonon ($\hat{H}_{\text{ph}}$) parts. Further, $\hat{H}_{\text{rest}} = \hat{H} - \hat{H}_{\text{ph}} - \left\langle \hat{H} - \hat{H}_{\text{ph}} \right\rangle_{\text{ph}}$ is the rest part of the interaction between the dressed exciton and new phonon. The symbol $\langle \cdot \rangle_{\text{ph}}$ denotes the averaging over new-phonon ensemble. Due to the fact that relaxation processes for the phonons which forms the thermal bath are rather fast (with the typical time constants in the biological macromolecular chains of order $10^{-14} - 10^{-11}$ s) in comparison with the characteristic time-scale of the transfer processes, they are populated according to an equilibrium statistical distribution. The explicit form of effective vibron Hamiltonian is

$$\hat{H}_{\text{vib}} = \mathcal{E} \sum_n a_n^\dagger a_n - J e^{-W(T)} \sum_n \left\{ a_n^\dagger a_{n-1} + a_n a_{n+1} \right\}$$

and the rest part of the Hamiltonian has the form

$$\hat{H}_{\text{rest}} = -J \sum_n \left\{ a_n^\dagger a_{n-1} \left( \theta_{n-1} - \langle \theta_{n-1} \rangle_{\text{ph}} \right) + a_n a_{n+1} \left( \theta_{n+1} - \langle \theta_{n+1} \rangle_{\text{ph}} \right) \right\} + \frac{1}{\sqrt N} \sum_{n,q} \left( F_q - \hbar \omega_q f_q^* \right) e^{i q n R_0} a_n^\dagger a_n (b_q + b_q^\dagger),$$

where $\langle \theta_{n+1} \rangle_{\text{ph}} = e^{-W(T)}$, $W(T) = N^{-1} \sum_q |f_q|^2 (2 \nu_q + 1)(1 - \cos(q R_0))$, is dressing fraction and $\nu_q = \left( e^{\hbar \omega_q / k_B T} - 1 \right)^{-1}$ describes the equilibrium population of the $q$-th phonon mode. The first term in (4) represents the fluctuations of the vibron-phonon interaction energy around its mean value. We assume that these fluctuations are small, and they can be disregarded. The second part is the main part of the polaron-phonon interaction which is responsible for the attenuation of the polaron amplitude.

We then transform our Hamiltonian from the coordinate to the wave vector $k$ representation using $a_n = 1/\sqrt N \sum_k e^{i k n R_0} a_k$, and $a_n^\dagger = 1/\sqrt N \sum_k e^{-i k n R_0} a_k^\dagger$ can be given as

$$\hat{H} = \hat{H}_{\text{vib}} + \hat{H}_{\text{ph}} + \hat{H}_{\text{rest}} = \sum_k \mathcal{E}_k a_k^\dagger a_k + \sum_q \hbar \omega_q b_q^\dagger b_q + \hat{H}_{\text{rest}},$$

where

$$\hat{H}_{\text{rest}} = \frac{1}{\sqrt N} \sum_{k,q} \left( F_q - \hbar \omega_q f_q^* \right) a_{k+q}^\dagger a_k (b_q + b_q^\dagger),$$

and

$$\mathcal{E}_k = \frac{1}{N} \sum_q \hbar \omega_q |f_q|^2 - \frac{1}{N} \sum_q F_q(f_q + f_q^*) - 2 J e^{-W(T)} \cos(k R_0)$$

is the polaron energy band measured from the vibron excitation energy. At this place, we will mention that the nature of self-trapped exciton state can be characterized by the mutual ratio of
three basic energy parameters: vibron energy band width $2J$, characteristic phonon energy $\hbar\omega_C$, and polaron binding energy $E_b = N^{-1} \sum_q |F_q|^2 / (\hbar\omega_q)$. In the process of vibron self-trapping in biological macromolecules, optical phonon modes are of particular significance [4, 15, 16]. For that reason, we restricted our calculations to the case of exciton interaction with non-dispersive optical phonon modes ($\omega_q = \omega_0 = \omega_C$). Standard small-polaron theory corresponds to such systems where $E_b \gg \hbar\omega_C \gg 2J$, i.e. to the systems with narrow energy band. In such structures, quasiparticle and phonon cloud forms new entity, dressed quasiparticle, and formed deformation instantaneously follows the motion of quasiparticle.

Further, in order to facilitate the analytic treatment, we assume that all phonon modes equally influence to exciton dressing. In this case, we introduce only one variational parameter $\delta$ ($0 < \delta < 1$), which can be defined as $f_q = \delta \cdot F_q^* / (\hbar\omega_q)$. This parameter measures the degree of the narrowing of the polaron band. Its value determines whether the vibron motion takes place in the coherent fashion (when $\delta$ takes low values) by means of the band mechanism, or it is achieved incoherently by the random jumps between neighbouring sites (for high values of $\delta$). Although it is technically less demanding, such an approach provides the results that are quite satisfying, especially for system parameter values that are of our interests [17].

Finally, the calculations are performed in terms of two system parameters, namely the adiabatic parameter ($B = 2J / (\hbar\omega_C)$) and the coupling constant ($S = E_b / (\hbar\omega_C)$). In this case, we have $W(\tau) = \delta^2 S \coth(1/2\tau)$, where $\tau = k_B T / \hbar\omega_0$ is the normalized temperature. Under these assumptions, the ground state polaron energy ($\mathcal{E}_{k=0}$) normalized to the characteristic phonon energy $\hbar\omega_0$ takes the form

$$E_{gs} = S\delta^2 (\delta - 2) - Be^{-S\delta^2 \coth(1/2\tau)}.$$  (8)

An optimal polaron state should be determined by the minimization of the polaron ground state energy, from the condition $\partial E_{gs} / \partial \delta = 0$. Consequently, the variational parameter can be found by solving of the following transcendental equation

$$\delta - 1 + \delta B \coth(1/2\tau)e^{-S\delta^2 \coth(1/2\tau)} = 0.$$  (9)

The dependence of the exciton dressing and its ground-state energy upon the basic system parameters and temperature are presented in Figs. 1 and 2.

![Figure 1](image_url)

**Figure 1.** The dependence of the vibron dressing (left) and the ground state energy (right) on the coupling constant for various values of the adiabatic parameter at temperature $T = 100$ K. Critical values of the adiabatic parameter $B_C = 0.68$ and the coupling constant $S_C = 1.3$ belong to moderately non-adiabatic and strong coupling limits.
Figure 2. The dependence of the vibron dressing (left) and the ground state energy (right) on the coupling constant for various values of the adiabatic parameter at temperature $T = 300$ K. Critical values of the adiabatic parameter $B_C = 0.22$ and the coupling constant $S_C = 0.4$ belong to non-adiabatic and weak coupling limits.

Obtained results appear to be consistent with the predictions of our earlier work concerned with the vibron self-trapping in the alpha-helix and two parallel chain structures [14, 18]. Briefly, we found that the system parameter space is divided into two characteristic areas: in the first area vibron dressing is a continuous function of the coupling constant $S$ for fixed values of $T$ and $B$. Such a behavior corresponds to the standard small-polaron quasiparticle. In the second area the vibron excitation abruptly changes its dressing: being initially a slightly dressed (practically free) quasiparticle, after an abrupt change its nature corresponds to the strongly dressed, practically immobile quasiparticle. The border between these two areas (determined by the critical values $B_C$ and $S_C$) depends on the temperature of the bath: with increasing of the temperature, its moves towards higher values of $S$ and $B$. It means that for the low temperatures the so called critical values belong to non-adiabatic ($B \ll 1$) and weak ($S \ll 1$) limits. However, for the room temperatures, the critical values may still belong to the non-adiabatic and intermediate-coupling! This result is interesting for understanding of the nature of the vibron dressing mechanism which is usually modelled in the framework of the adiabatic (soliton) polaron picture [1, 2, 3, 19]. On the other hand, it is consistent with some numerical investigations provided by Hamm and Tsironis [20].

3. Finding of the polaron amplitude damping

Since the characteristic time-scale of the exciton transfer processes is much larger than the time of phonon relaxation, in order to describe exciton migration processes it is necessary to use the non-equilibrium statistical distribution for exciton subsystem, i.e. the density matrix of excitons must be described by non-equilibrium density operator. On the other hand, as it was mentioned, the phonon subsystem is regarded as a subsystem in the equilibrium state and it is described by the equilibrium statistical operator. Consequently, in order to examine the polaron damping, we follow the well-known approach developed within the theory of the open quantum systems [21, 22], and we derive a set of evolution equations for non-equilibrium statistical average values $\langle a_k \rangle = \text{Tr} \langle \rho(t) a_k \rangle$ of polaron operators $a_k$. Here $\rho(t) = \rho_e(t) \otimes \rho_{ph}$ is the time dependent non-equilibrium statistical operator (whereas $\rho_e(t)$ is the non-equilibrium time-dependent exciton statistical operator, while $\rho_{ph}$ is phonon statistical operator). Evolution of the mean value of operator $A$ is determined by the relation $i\hbar \langle \dot{A} \rangle / dt = \langle [A, H] \rangle$. For the Hamiltonian (5) we...
obtain
\[ i\hbar \frac{d}{dt} \langle a_k \rangle = \mathcal{E}_k \langle a_k \rangle + \sum_q \phi_q \left\{ \langle a_{k-q}b_q \rangle + \langle a_{k-q}b_q^\dagger \rangle \right\}, \]

(10)

where \( \phi_q = (F_q - \hbar \omega_q f_q^*)/\sqrt{N} \) (in the case of \( \delta \) variational approach this parameter becomes \( \phi_q = F_q(1-\delta)/\sqrt{N} \)). In the equation above, there appear two new amplitudes, namely \( \langle a_{k-q}b_q \rangle \) and \( \langle a_{k-q}b_q^\dagger \rangle \). The equation of motion for these amplitudes contain other new amplitudes: \( \langle a_k b_{q_1} b_{q_2} \rangle, \langle a_k b_{q_1}^\dagger b_{q_2} \rangle, \langle a_{k_1} a_{k_2} a_{k_3} \rangle, \langle a_k b_{q_1} b_{q_2}^\dagger \rangle, \) and \( \langle a_k b_{q_1}^\dagger b_{q_2}^\dagger \rangle \). The equation of motion for these amplitudes contain amplitudes that correspond to multiparticle processes whose probability is lower in order of magnitude than the probability of single-particle processes. For this reason, it is possible to cut-off the above mentioned amplitudes as follows: \( \langle a_k b_{q_1} b_{q_2} \rangle = \langle a_k \rangle \langle b_{q_1} b_{q_2} \rangle = 0, \)

\( \langle a_k b_{q_1}^\dagger b_{q_2} \rangle = \langle a_k \rangle \langle b_{q_1}^\dagger b_{q_2} \rangle = 0, \)

\( \langle a_k b_{q_1} b_{q_2}^\dagger \rangle = \langle a_k \rangle \langle b_{q_1} b_{q_2}^\dagger \rangle = \langle a_k \rangle \nu_q \delta_{q_1,q_2}, \)

\( \langle a_k b_{q_1}^\dagger b_{q_2}^\dagger \rangle = \langle a_k \rangle (1 + \nu_q) \delta_{q_1,q_2}. \)

One should have in mind that the phonon subsystem is in the equilibrium state. Finally, we neglect all the multipolaron processes \( \langle a_{k_1} a_{k_2}^\dagger \rangle \approx 0. \)

Under these assumptions the equations of motion for \( \langle a_{k-q}b_q \rangle, \) and \( \langle a_{k-q}b_q^\dagger \rangle \) take the form:

\[ i\hbar \frac{d}{dt} \langle a_{k-q}b_q \rangle = (\mathcal{E}_{k-q} + \hbar \omega_q) \langle a_{k-q}b_q \rangle + \phi_{-q} \nu_q + 1) \langle a_k \rangle, \]

(11)

\[ i\hbar \frac{d}{dt} \langle a_{k-q}b_q^\dagger \rangle = (\mathcal{E}_{k-q} - \hbar \omega_q) \langle a_{k-q}b_q^\dagger \rangle + \phi_{-q} \nu_q \langle a_k \rangle. \]

(12)

After integration of Eqs. (11) and (12), one can obtain

\[ \langle a_{k-q}b_q \rangle = -\langle a_k \rangle \left\{ \mathcal{P} \left( \frac{1}{\mathcal{E}_{k-q} + \hbar \omega_q} \right) + i\pi \delta(\mathcal{E}_{k-q} + \hbar \omega_q) \right\} \phi_{-q}(\nu_q + 1), \]

(13)

\[ \langle a_{k-q}b_q^\dagger \rangle = -\langle a_k \rangle \left\{ \mathcal{P} \left( \frac{1}{\mathcal{E}_{k-q} - \hbar \omega_q} \right) - i\pi \delta(\mathcal{E}_{k-q} - \hbar \omega_q) \right\} \phi_{-q} \nu_q, \]

(14)

where \( \mathcal{P} (1/|\mathcal{E}_k \pm \hbar \omega_q|) \) are the principal values of corresponding integrals. After substitution of Eqs. (13), (14) into Eq. (10), we obtain a Schrödinger-like equation with damping

\[ i\hbar \frac{d}{dt} \langle a_k \rangle = (\mathcal{E}_k - \Delta_k) \langle a_k \rangle - i\gamma_k \langle a_k \rangle \]

(15)

Such an equation describes the non-equilibrium dynamics of polaron, and it determines the polaron energy shift and polaron amplitude damping caused by the polaron interaction with the environment (i.e. with the thermal oscillations of the macromolecular chain). Here

\[ \Delta_k = \sum_q |\phi_q|^2 \nu_q \mathcal{P} \left( \frac{1}{\mathcal{E}_{k+q} - \hbar \omega_q} \right) + \sum_q |\phi_q|^2 (1 + \nu_q) \mathcal{P} \left( \frac{1}{\mathcal{E}_{k+q} + \hbar \omega_q} \right) \]

(16)

is the shift of the polaron energy, and

\[ \gamma_k = \pi \left\{ \sum_q |\phi_q|^2 \nu_q \delta(\mathcal{E}_{k+q} - \hbar \omega_q) + \sum_q |\phi_q|^2 (1 + \nu_q) \delta(\mathcal{E}_{k+q} + \hbar \omega_q) \right\} \]

(17)

is the damping factor of the polaron amplitude which determines the characteristic relaxation time \( \tau_k \sim 1/\gamma_k \). As can be remarked, the polaron amplitude damping is caused by the emission
and absorption of real phonons, because the presence of the delta-functions in $\gamma_k$ shows that the energy is conserved during such processes. On the other hand, the shift of the polaron energy $\Delta_k$ arises as a consequence of the emission and absorption of virtual phonons, because it is meaningful only in the case when $E_{k+q} \neq \pm \hbar \omega_q$.

In the case when the vibron interacts with non-dispersive optical phonon modes the normalized energy of damped system looks as

$$\Delta_k = S(1 - \delta)^2 I_\Delta,$$

where integral $I_\Delta$ has the form

$$I_\Delta = \frac{1}{N} \sum_q \cot(1/2 \tau) \left[ S \delta(\delta - 2) - Be^{-S\delta^2 \cot(1/2 \tau)} \cos (k - q) R_0 \right] - 1,$$

and the damping factor becomes

$$\gamma_k = \pi E_b (1 - \delta)^2 I_\gamma,$$

where

$$I_\gamma = \frac{e^{1/\tau}}{e^{1/\tau} - 1} \frac{1}{N} \sum_q \delta(\bar{E}_{k-q} + 1) + \frac{1}{e^{1/\tau} - 1} \frac{1}{N} \sum_q \delta(\bar{E}_{k-q} - 1).$$

The dependence of the damping factor on the system parameters and temperature is shown in Fig. 3.

**Figure 3.** The dependence of the vibron damping on the coupling constant for various values of the adiabatic parameter at temperatures $T = 100$ K (left) and $T = 300$ K (right).

From the above results, one can remark that the damping of the vibron amplitudes induced by the residual polaron-phonon interaction is significant in the area of the system parameter space where low or partial vibron dressing takes place (i.e., where $\delta < 1$). In the system parameter area where the exciton is slightly dressed, the amplitude damping rises with the increasing of $S$. But in the area where the exciton becomes heavy dressed, $\gamma$ sharply decreases. Such a behavior is much more expressed for high $B$ (i.e., when the system becomes more adiabatic). Additionally, this tendency increases with the rise of $B$, and for the system parameters where the dressing abruptly changes, the damping becomes also discontinuous. The maximal value of $\gamma$ increases with increasing of $B$ and $T$.

Obtained results suggest that the increasing of system temperature can drive the biological MCs to the system parameter area where the polaron crossover is possible. In this area of
the parameter space, the systems are quite sensitive to small variations of temperature and the values of other system parameters. Amplitude damping of slightly dressed vibrons (caused by residual vibron-phonon interaction) in (moderately) non-adiabatic region strongly increases with the increasing of the coupling constant and temperature of the thermal bath. In the nearly adiabatic and adiabatic region such increasing is noticeably lower. At the same time, the residual vibron-phonon interaction practically has no influence on the amplitude damping of heavy dressed vibrons. The mean lifetime of nearly free vibrons is very short (as compared to the mean lifetime in the low temperature limit), and consequently the width of vibron spectral lines of the absorption spectra should be high. These results could explain the absence of observation of weak shift of the anomalous peak in the absorption spectra of crystalline acetanilide, which was predicted by the use of the partially dressed SP picture.

Acknowledgments

This work was supported by the Serbian Ministry of Education and Science under Contract Nos. III-45010, III-45005 and by the Project within the Cooperation Agreement between the JINR, Dubna, Russian Federation and Ministry of Education and Science of Republic of Serbia. One of the authors (A.Ch.) thanks the Blokhintsev-Votruba Program (JINR, Dubna) for the financial support of participation in the conference ISQS25.

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