Coherent population trapping in quantized light field

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A full quantum treatment of coherent population trapping (CPT) is given for a system of resonantly coupled atoms and electromagnetic field. We develop a regular analytical method of the construction of generalized dark states (GDS). It turns out that GDS do exist for all optical transitions \( F_g \rightarrow F_e \), including bright transitions \( F \rightarrow F + 1 \) and \( F'' \rightarrow F'' \) with \( F'' \) a half-integer, for which the CPT effect is absent in a classical field.

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Coherent population trapping (CPT) for atoms interacting with a resonant field is now well-known and widely used in various branches of atomic and laser physics. Let us list several examples: nonlinear optics of resonant media, nonlinear high-resolution spectroscopy, high-precision metrology (atomic clocks and magnetometers), laser cooling, atom optics and interferometry, and quantum information processing. Originally the CPT theory was developed for a three-state model. A generalization to multilevel atomic systems was done by Hioe and Carroll without regard for the relaxation. In particular, in sufficient conditions for the existence of dark (non-coupled with light) states at the two-photon resonance have been derived. For the resonant excitation by a classical elliptically polarized field from the atomic ground state with angular-momentum degeneracy, necessary and sufficient conditions for the occurrence of CPT and the explicit form of dark states have been established in our works. With allowance made for the radiative relaxation, all optical transitions \( F_g \rightarrow F_e \) \( (F_g, F_e) \) are the total angular momenta of the ground (g) and excited (e) states were classified into two classes depending on the occurrence of CPT: (i) dark transitions \( F \rightarrow F - 1 \) with \( F \) arbitrary and \( F'' \rightarrow F'' \) with \( F'' \) an integer; (ii) bright transitions \( F \rightarrow F + 1 \) with \( F \) arbitrary and \( F'' \rightarrow F'' \) with \( F'' \) a half-integer. In the latter case CPT is absent.

It was shown in that the laser-field fluctuations of certain type destroy the dark state, and the CPT effect disappears. This rule, however, is not general. For instance, we have found in that the dark state can arise upon the interaction with an arbitrarily fluctuating monochromatic radiation field. In particular, it has been shown that the CPT effect occurs even if the photon spin coherence is completely absent.

Recent outstanding theoretical works on the problem of dark-state polariton and the experimental demonstration of the optical information storage in resonant atomic media attracted an attention to the problem on the most general form of dark state in the “atoms+field” system. It should be noted that the experimental results obtained in are of great importance in view of their possible applications to the storage of essentially quantum information encoded in nonclassical states of light. In this connection, the following statement of the problem looks quite reasonable: what happens with the CPT existence conditions and with the form of dark states, when the resonant field is described by the creation and annihilation operators instead of c-number amplitudes? From this point of view the dark-state polariton should be a particular case of the general solution, when one field mode is classical (i.e. it is in a coherent state), while another mode is quantal. Moreover, as it was argued in, it is quite possible that, for the long-term storage of quantum information in an atomic medium, it will be necessary to use ultracold atomic ensembles (e.g. Bose-Einstein condensates). In this case, atoms can conveniently be described by the secondary quantized amplitudes, i.e. by the creation and annihilation operators of Bose or Fermi types.

In the present paper coherent population trapping in an atomic ensemble with optical transition \( F_g \rightarrow F_e \) upon the resonance interaction with a quantized monochromatic running wave is considered. We develop a regular method of the construction of generalized dark states (GDS) for arbitrary angular momenta \( F_g, F_e \). It is shown that for the dark transitions \( (F \rightarrow F - 1) \) with \( F \) arbitrary and \( F'' \rightarrow F'' \) with \( F'' \) an integer the CPT effect occurs at arbitrary numbers of atoms and photons, representing a direct generalization of the results obtained for the classical elliptically polarized field. Unexpectedly, it turns out that in the quantized field CPT can appear even for the bright transitions \( (F \rightarrow F + 1) \) with \( F \) arbitrary and \( F'' \rightarrow F'' \) with \( F'' \) a half-integer, for which the dark states are absent in the classical field. In this case, however, we have certain limitations on the number of photons in one of two polarization modes for the transitions \( F'' \rightarrow F'' \) with \( F'' \) a half-integer, and on the numbers of both atoms and photons for the transitions \( F \rightarrow F + 1 \) with \( F \) arbitrary.

Hereinafter, we assume that atoms obey to either Bose-Einstein or Fermi-Dirac statistics. The total Hamiltonian of the “atoms+field” system is a sum of three summands:

\[
\hat{H} = \hat{H}_a + \hat{H}_{ph} + \hat{H}_{a-ph}.
\]

The Hamiltonian of free atoms can be written as

\[
\hat{H}_a = \hbar \omega_0 \sum_{\mu} c_{\mu}^\dagger c_{\mu}, \quad (1)
\]
where $\omega_0$ is the optical transition frequency, $\hat{c}_{\mu s}^\dagger$ and $\hat{c}_{\mu s}$ are the operators of creation and annihilation of an atom in the excited-state Zeeman substate $|F_e, \mu_e\rangle \ (-F_e \leq \mu_e \leq F_e)$. If the quantization axis is directed along the wavevector, then the Hamiltonian of free transversal photons reads

$$\hat{H}_{ph} = \hbar \omega \sum_{s = \pm 1} \hat{a}_s\hat{a}_s,$$  

(2)

where $\omega$ is the field frequency, $\hat{a}_s (\hat{a}_s^\dagger)$ is the bosonic annihilation (creation) operator of a photon with the right ($s = +1$) or left ($s = -1$) circular polarization. In the rotating wave approximation the dipole coupling transversal photons reads

$$V = \hbar \Omega \sum_{\mu_{g,s}} \hat{c}_{\mu g,s}^\dagger \hat{a}_s.$$

(3)

Here $\Omega$ is the single-photon Rabi frequency, $C_{\alpha_F g, \mu_g, 1s}$ are the Clebsch-Gordan coefficients, and $\hat{b}_{\mu g}^\dagger (\hat{b}_{\mu g})$ is the operator of creation (annihilation) of an atom in the ground-state Zeeman substate $|F_g, \mu_g\rangle \ (-F_g \leq \mu_g \leq F_g)$.

Our goal is to find the generalized dark states $|NC\rangle$ of the system “atoms-field”, which nullify the interaction operator:

$$\hat{V} |NC\rangle = 0.$$  

(4)

In addition the state $|NC\rangle$ should be stable with respect to the radiative relaxation, i.e. GDS should not contain atoms in the excited state. Formally this condition can be expressed as

$$\hat{H}_a |NC\rangle = 0,$$  

(5)

in line with eq.(1), where just the excited-state operators $\hat{c}_{\mu s}$ are present. Combining eq.(4) and eq.(5), one can see that $\hat{H}_{a-ph} |NC\rangle = 0$.

Taking into account the angular-momentum selection rules, we see that the generic scheme of the light-induced transitions is split into two independent chains for arbitrary optical transition $F_g \rightarrow F_e$. There exist three types of such chains $\Lambda$. For the first $\Lambda$-type of chains the number of coupled ground-state substates is greater than those in the excited state by one. The $\Lambda$-chains are realized (see in Fig.1a,b) in the transitions $F \rightarrow F - 1$ with $F$ arbitrary and $F' \rightarrow F'$ with $F'$ an integer. For the second $V$-type of chains the number of coupled ground-state substates is less than those in the excited state by one. The $V$-chains are realized (see in Fig.1b,c) for the transitions $F \rightarrow F + 1$ with $F$ arbitrary and $F' \rightarrow F'$ with $F'$ an integer. For the third type of chains the number of coupled ground-state substates is equal to those in the excited state. These chains will be referred to as $N$-chains. The $N$-chains are realized (see in Fig.1d) for the transitions $F'' \rightarrow F''$ with $F''$ a half-integer. Let us develop a procedure of the GDS construction for each type of chains separately.

The $\Lambda$-chains. Consider the $\Lambda$-chain consisting of $L$ links. For the sake of convenience we re-enumerate the substates as it is shown in Fig.1a (lower panel). The ground-state substates have odd numbers, while those in the excited state have even numbers. The coupling operator projection on this $\Lambda$-chain sub-space can be written as

$$\hat{V}_{\Lambda} = \hbar \Omega \sum_{j=1}^L \hat{c}_{2j}^\dagger (G_{2j-1}^{2j} \hat{b}_{2j-1} \hat{a}_{j+1} + G_{2j+1}^{2j} \hat{b}_{2j+1} \hat{a}_{j-1}),$$  

(6)

where $G_k^q$ denote the corresponding Clebsch-Gordan coefficients (compare with eq.(3)).

We will need in the following construction:

$$\hat{\Psi}_{NC} = \sum_{j=0}^L \hat{A}_{2j+1} \hat{b}_{2j+1}^\dagger,$$  

(7)

which is a superposition of the atomic ground-state creation operators with operator coefficients $\hat{A}_{2j+1}$. These coefficients are functions of the photon annihilation operators $\hat{a}\pm$.

Using the standard commutation (bosons) or anticommutation (fermions) rules, one can obtain:

$$\hat{V}_{\Lambda} \hat{\Psi}_{NC} = \pm \hat{\Psi}_{NC} \hat{V}_{\Lambda} + \hbar \Omega \sum_{j=1}^L \hat{c}_{2j}^\dagger (G_{2j-1}^{2j} \hat{a}_{j+1} \hat{A}_{2j+1} + G_{2j+1}^{2j} \hat{a}_{j-1} \hat{A}_{2j+1}),$$  

(8)

where the sign $+/-$ corresponds to bosons/fermions. The coefficients $\hat{A}_l$ are chosen in such a way that the second term in the r.h.s. of eq.(5) becomes zero. In the other word $\hat{A}_l$ obey the recurrent operator equations:

$$G_{2j-1}^{2j} \hat{a}_{j+1} \hat{A}_{2j+1} + G_{2j+1}^{2j} \hat{a}_{j-1} \hat{A}_{2j+1} = 0.$$  

(9)

These equations always have a solution, because the number of equations in eq.(9) equals $L$, while the number of the coefficients $\hat{A}_k$ equals $L + 1$. The solution can be written in the form of positive powers of the annihilation operators $\hat{a}\pm$:

$$\hat{A}_{2j+1} = (-1)^j (\hat{a}_{j+1})^j (\hat{a}_{j-1})^{L-j} \left[ \prod_{q=1}^j G_{2q-1}^{2q} \right] \left[ \prod_{q=j+1}^L G_{2q+1}^{2q} \right],$$  

(10)

where $j = 0, \ldots, L$, and the convention $\prod_{k=1}^k 1 = 1$ is used. With this choice of the coefficients $\hat{A}_{2j+1}$ it follows from eq.(9) that

$$\hat{V}_{\Lambda} \hat{\Psi}_{NC} = \pm \hat{\Psi}_{NC} \hat{V}_{\Lambda}.$$  

(11)

This fundamental relationship allows us to construct the dark state $|NC\rangle_\Lambda$ for the $\Lambda$-chain in the form:

$$|NC\rangle_\Lambda = (\hat{\Psi}_{NC})^n \hat{\Psi}(\hat{a}^\dagger) |0\rangle,$$  

(12)
where $n$ is arbitrary non-negative number, the functional \( \tilde{\Phi}\{\hat{a}^\dagger\} \) depends exclusively on the photon operators \( \hat{a}_k^\dagger \), and \( |0\rangle \) is the vacuum state of atoms and photons. Indeed, using eq. (11), we get

\[
\hat{V}_\Lambda |NC\rangle_\Lambda = (\pm 1)^n (\tilde{\Psi}_{NC})^n \hat{V}_\Lambda \tilde{\Phi}\{\hat{a}^\dagger\} |0\rangle = 0.
\] (13)

The later equality follows from the fact that the coupling operator \( \hat{V}_\Lambda \) contains the annihilation operators \( b_k \), which commute with \( \tilde{\Phi}\{\hat{a}^\dagger\} \) and give zero, acting on the vacuum state. It is also obvious that the state \( |NC\rangle_\Lambda \) obeys the equation (5), because the excited-state operators \( c_k^\dagger \) do not enter in the construction (7).

The exponent \( n \) in eq. (12) means the number of atoms participating in the formation of the dark state. In the Bose case this number can be arbitrary, while in the Fermi case we have just two possibilities \( n = 0, 1 \), since \( (\tilde{\Psi}_{NC})^2 = 0 \). However, this fact does not lead to the principal limitation on the number of atoms in an ensemble. Indeed, taking into account the translational degrees of freedom, for example in the momentum representation, we can write the more general form of GDS:

\[
|NC\rangle_\Lambda = \prod_p (\tilde{\Psi}_{NC}(p))^{n_p} \tilde{\Phi}\{\hat{a}^\dagger\} |0\rangle,
\] (14)

where the creation operators \( \hat{b}_k^\dagger(p) \) and the basic construction \( \tilde{\Psi}_{NC}(p) \) are labeled by the momentum \( p \). The total number of atoms is \( \sum_p n_p \), and it can be arbitrary in the Fermi case as well. It should be noted that an arbitrary superposition of the dark states (14) will be dark, according to our definitions (4) and (6).

It is interesting that the basic construction \( \tilde{\Psi}_{NC} \) can be obtained from the results of our early work (15) in a formal way. To do this one has to use the explicit form of the dark states in the classical field, substituting formally the Zeeman state wavefunctions by the atom creation operators \( \hat{b}_k^\dagger \) and the circular field components by the photon annihilation operators \( \hat{a}_k \). In this sense the obtained GDS \( |NC\rangle_\Lambda \) can be viewed as a direct generalization of the results of the paper (12) to the quantized field and many-particle atomic system.

As an example, we demonstrate here how does the \( m \)-fold excited dark-state polariton \( |\Lambda\rangle \) emerge in our approach. We consider the simplest \( \Lambda \)-chain (i.e. \( L = 1 \)) with the operator construction (7) given by \( \hat{V}_{NC} = G_2 \hat{a}_{-1}^\dagger \hat{b}_1^\dagger - G_3 \hat{b}_{-1}^\dagger \hat{a}_1 \). The functional \( \tilde{\Phi}\{\hat{a}^\dagger\} \) in eq. (12) is taken in the form:

\[
\tilde{\Phi}\{\hat{a}^\dagger\} = N_0 (\hat{a}_{-1}^\dagger)^m \exp(Z \hat{a}_{-1}^\dagger),
\] (15)

where \( N_0 \) is a normalization constant. When acting on the vacuum state \( |0\rangle \), the operator (15) generates a coherent state with amplitude \( Z \) in the mode with circular polarization \(+1\) and \( m \) photons in the mode with circular polarization \( -1 \). The direct calculations by formula (12) with \( G_2^2 = G_3^2 \) yield the result that coincides (except for notation) with the \( m \)-fold excited state \( |D, m\rangle \) of the dark-state polariton \( \Lambda \).

The \( N \)-chains. Consider the Zeeman substates, for which the light-induced transitions form the \( N \)-chains, as it is shown in Fig. 1d (two lower panels) for the transitions \( F'' \to F'' \) of half-integer. Then the coupling operator projections on these subspaces are written as

\[
\hat{V}_{N_+} = \hat{V}_\Lambda + h\Omega G_{2L+1}^{2L+2} c_1^\dagger \hat{a}_{-1}^\dagger b_{2L+1},
\] (16)

\[
\hat{V}_{N_-} = \hat{V}_\Lambda + h\Omega G_{2L}^2 c_1^\dagger \hat{a}_{-1}^\dagger b_1,
\] (17)

where \( \hat{V}_\Lambda \) denotes the part of the interaction operator connecting the substates into the maximal \( \Lambda \)-chain with \( L \) links. This \( \Lambda \)-chain is marked in Fig. 1d by double lines. The explicit form of \( \hat{V}_\Lambda \) corresponds to eq. (5). The second terms in the operators (16) and (17) describe the interaction via the outermost excited-state Zeeman substate, which is connected with just one ground-state substate. This coupling is shown in Fig. 1d by single line.

For the main contribution \( \hat{V}_\Lambda \), in line with eq. (14) and eq. (10), we construct the operator \( \tilde{\Psi}_{NC} \), which obeys the equation (11). Then, for the \( N \)-chain the dark state \( |NC\rangle_{N_+} \), obeying the equation \( \hat{V}_{N_+} |NC\rangle_{N_+} = 0 \), has the form:

\[
|NC\rangle_{N_+} = (\tilde{\Psi}_{NC})^n (\hat{a}_{-1}^\dagger)^m \tilde{\Phi}\{\hat{a}_{-1}^\dagger\} |0\rangle, \quad (m \leq L),
\] (18)

where the generic operator functional \( \tilde{\Phi}\{\hat{a}_{-1}^\dagger\} \) depends only on the left-polarized photon creation operators \( \hat{a}_{-1}^\dagger \). Thus, the number of the left-polarized photon can be arbitrary. It is obvious that (18) nullifies the operator \( \hat{V}_\Lambda \) (see eq. (13)). The limitation on the number of the right-polarized photons \( m \) in eq. (18) is connected with the necessity to nullify the additional contribution \( h\Omega G_{2L+1}^{2L+2} c_1^\dagger \hat{a}_{-1}^\dagger b_{2L+1} \) in r.h.s. of eq. (16). The condition \( m \leq L \) follows from the fact that the operator construction \( \tilde{\Psi}_{NC} \) contains the creation operator \( \hat{b}_{2L+1}^\dagger \) with the coefficient \( A_{2L+1} \propto \hat{a}_{-1}^\dagger \) (see eq. (12)).

In much the same way, we find for the \( N_- \)-chain:

\[
|NC\rangle_{N_-} = (\tilde{\Psi}_{NC})^n (\hat{a}_{-1}^\dagger)^m \tilde{\Phi}\{\hat{a}_{+1}^\dagger\} |0\rangle, \quad (m \leq L),
\] (19)

where the number of the left-polarized photons is limited by \( m \leq L \).

Strictly speaking, in eqs. (18), (19) we have to put the number of atoms \( n = 0, 1 \), since the \( N \)-chains correspond to half-integer angular momenta, i.e. to fermions. Because of this, analogously to (14), we present the dark state in more general form:

\[
|NC\rangle_{N_{\pm}} = \left( \prod_p (\tilde{\Psi}_{NC}(p))^{n_p} \right) (\hat{a}_{\pm1}^\dagger)^m \tilde{\Phi}\{\hat{a}_{\pm1}^\dagger\} |0\rangle.
\] (20)

Now the number of atoms \( \sum_p n_p \) can be arbitrary.

Evidently, for the \( N \)-chains the \( m \)-fold excited dark-state polariton \( |D, m\rangle \) can be constructed. However,
contrary to the $\Lambda$-chains, here the number of the quantized mode excitations is limited by the length of chain ($m \leq L$).

The $V$-chains. Consider the Zeeman substates, for which the light-induced transitions form the $V$-chain, as it is shown in Fig.1c (lower panel). Then the coupling operator projection on this subspace can be written as

$$\hat{V}_V = \hat{V}_\Lambda + \mathcal{H}(G_{1G}^{0}\hat{b}_1 - G_{1G}^{2L+2}\hat{b}_{2L+1} + G_{2L+1}\hat{b}_{2L+2} \hat{b}_1), \tag{21}$$

where $\hat{V}_\Lambda$ denotes the part of the interaction operator connecting the substates into the maximal $\Lambda$-chain with $L$ links. This $\Lambda$-chain is marked in Fig.1c by double lines. The explicit form of $\hat{V}_\Lambda$ corresponds to eq. 9. The two additional terms in the operator $\hat{V}_\Lambda$ describe the interaction via the outermost excited-state Zeeman substates, which are connected with just one ground-state substate. This coupling is shown in Fig.1c by single line.

Again, for the main contribution $\hat{V}_\Lambda$, in line with eq.17 and eq.10, we construct the operator $\hat{\Psi}_{NC}$, which obeys the equation 14. Then, for the $V$-chain the dark state can be written in the form:

$$|NC\rangle_V = \hat{\Psi}_{NC}(\hat{a}_1^\dagger)^m(\hat{a}_{-1}^\dagger)^{m'}|0\rangle, \quad (m, m' \leq L). \tag{22}$$

Here, contrary to the $N$-chains, the numbers of photons of both polarizations are limited ($m, m' \leq L$). This is because we have to nullify the two additional terms in r.h.s. of eq.21. Furthermore, the state 22 is non-trivial (i.e. it differs from zero and contains photons) if and only if $(m + m') > L$.

As an example, we consider the $V$-chain with one $\Lambda$-link, i.e. $L = 1$. In this case $\hat{\Psi}_{NC} = G_{1G}^{2}\hat{a}_{-1}^\dagger \hat{b}_1^\dagger - G_{1G}^{2L+1}\hat{a}_1^\dagger \hat{b}_{2L+1}^\dagger$, and $m = m' = 1$. Then using eq.22, we obtain the expression:

$$|NC\rangle_V = (G_{1G}^{2}\hat{a}_{-1}^\dagger \hat{b}_1^\dagger - G_{1G}^{2L+1}\hat{a}_1^\dagger \hat{b}_{2L+1}^\dagger)|0\rangle,$$

where the atom and field variables are entangled. Here the quantum entanglement is precisely the reason leading to the non-trivial dark states on the bright optical transitions.

It should be noted that the dark states 22 are realized for just one atom. It can be seen from the calculation:

$$(\hat{\Psi}_{NC})^n(\hat{a}_{-1}^\dagger)^m(\hat{a}_1^\dagger)^{m'}|0\rangle = 0, \quad (n \geq 3; m, m' \leq L).$$

This equality follows from the simple count the numbers of creation and annihilation operators in each term. In the case of two atoms the state $(\hat{\Psi}_{NC})^2(\hat{a}_1^\dagger)^m(\hat{a}_{-1}^\dagger)^{m'}|0\rangle$ is trivial or (at $m = m' = L$) it does not contain photons.

Concluding, we have developed the analytical method, which allows us to construct a wide class of GDS for the chains of $\Lambda, V, N$-types. These chains of the dipole connected Zeeman substates appear under the consideration of the resonance interaction of the quantized plane-wave modes with optical transitions $F'_g \rightarrow F'_e$. The generalized dark states exist for arbitrary field frequency $\omega$, they immune to the radiative relaxation, and their mathematical structure corresponds, in general case, to quantum states entangled in atomic and field variables (see eqs. 7, 11). The obtained results lead to the following classification of the optical transitions. (i) The transitions $F \rightarrow F - 1$ with arbitrary $F$ and $F' \rightarrow F'$ with $F'$ an integer, where the $\Lambda$-chains are realized. Here GDS exist for arbitrary quantum state of the field and for arbitrary number of atoms. (ii) The transitions $F'' \rightarrow F''$ with $F''$ a half-integer, where the $N$-chains are realized. In this case GDS occur at arbitrary number of atoms, but the number of photons of the right (or left) circular polarization is limited. (iii) Transitions $v \rightarrow F + 1$, where the $V$-chains are realized. Here we found just the ultra-quantum GDS for one atom interacting with the limited numbers of photons of both right and left polarizations.

We would like to stress that in the cases (ii) and (iii) the ground-state dark states are absent in the classical light field. It is possible that the obtained GDS do not exhaust all dark states in the “atoms+field” system. In particular, the problem on the existence of a many-particle dark state for the transitions $F \rightarrow F + 1$ is still open. Note also that all the explicit expressions for GDS can be easily re-formulated for distinguishable particles. To do this one has to use the Zeeman wavefunctions instead of the creation operators $\hat{b}_{k}^\dagger$ in the basic construction 9.

We have found the generalized dark states for the given frequency $\omega$ and wave-vector $k$, then these states can be marked by $\omega$ and $k$: $|NC(\omega, k)\rangle$. It is obvious that an arbitrary superposition and even an incoherent mixture of the dark states corresponding to different $\omega$ and $k$ will obey the conditions 11 and 14, i.e. they will be dark states.

The obtained results are of principal significance for the CPT theory, quantum optics, and quantum informatics in resonant atomic media.

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FIG. 1: The light-induced transitions between Zeeman substates in a resonant plane wave: 
a) transitions \( F \rightarrow F - 1 \) with \( 2 \rightarrow 1 \) as an example (upper panel), the \( \Lambda \)-type chain (lower panel); 
b) transitions \( F' \rightarrow F' \) (\( F' \) is an integer) with \( 2 \rightarrow 2 \) as an example; 
c) transitions \( F \rightarrow F + 1 \) with \( 1 \rightarrow 2 \) as an example (upper panel), the \( V \)-type chain (lower panel); 
d) transitions \( F'' \rightarrow F'' \) (\( F'' \) is a half-integer) with \( 3/2 \rightarrow 3/2 \) as an example (upper panel), the \( N_{\pm} \)-chains (two lower panels).