Quantum control of exciton states in clusters of resonantly interacting fluorescent particles using biharmonic laser pumping

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The quantum control of dimers, trimers, and tetramers of resonantly interacting fluorescent particles using a biharmonic laser pumping is analyzed. Special emphasis is given on the preparation of all possible pure exciton states (e.g., for tetramers these are single-, two-, three-, and four-exciton states) and their maximally entangled Bell states. The general results are illustrated using as an example the pair and quartet centers of neodymium ions in calcium fluoride (*M* and *N* centers), where all necessary experimental information concerning the interactions and decoherence is available, and the experimental preparation of Bell vacuum-single exciton and vacuum-biexciton states have been recently demonstrated. These results can be easily rescaled for the cases of quantum dots and dye molecules. We show that the broad set of quantum-logic operations under such systems can be effectively performed, which again confirms their applicability as prospective quantum computer hardware. Numerical results are compared with the analytical results obtained for a particular case of the biharmonic excitation of dimers. Excellent agreement between these approaches is demonstrated.

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I. INTRODUCTION

On-demand laser-assisted quantum manipulations under systems of resonantly interacting fluorescent particles and their use for quantum informatics and related topics currently attract a lot of attention (see, for example, Refs. 1–21). Quite recently the first experiments reporting the preparation of Bell-entangled states in such systems composed by quantum dots,[4–8] dye molecules,[9,10] and dopant rare-earth ions,[11] and the performance of different quantum manipulations upon them were reported, which makes detailed theoretical consideration of these systems especially timely. Despite a lot of efforts in this direction (amongst the cited references, see especially Refs. 1–3 and 12–18), important aspects of the problem still remain to be studied.

First, it should be noted that almost all researches restrict themselves with the consideration of only the single frequency, i.e., monochromatic, (pulsed) optical action upon the system at hand. Understandable as this might seem, thanks to the existence of the well-elaborated and ready-to-use rotating-wave approximation suitable to theoretically treat exactly this problem, in practice the simultaneous action of laser pulses with different frequencies upon the systems of resonantly interacting fluorescent particles is needed. Only such a "parallel action" enables us to drastically increase the number of quantum operations performed under the system during the decoherence time thus much improving its applicability for the quantum computing. It enables us also to essentially decrease the intensity of laser pulses applied (and hence to decrease the laser heating and its undesirable consequences); for example, our calculations show that one needs a hundred-times-less intensive pulses to prepare the Bell vacuum-biexciton state for the pair of fluorescent particles when using biharmonic pumping instead of a single-frequency one (see below). Second, in almost all papers only rather specific types of entanglement, that is entanglement between the vacuum and the "highest-excited" state of the system (e.g., three exciton for trimer or four exciton for tetramer) and its closest analogues are analyzed. This also seems understandable, because such states are nothing else than a kind of celebre Greenberger-Horne-Zeilinger state[22] and, what we believe is especially important in the current context, they also can be efficiently prepared via a single-frequency laser pumping. At the same time, other interesting and no less appealing in their potential applicability for quantum computing nonlocal entangled states can be prepared: we mean, e.g., entangled vacuum–single-exciton states for trimers, vacuum–two-exciton states for tetramers, and so on. What is very important, the preparation of such relatively low-energy states, again requires less-intensive laser radiation (especially when a biharmonic pumping is used), which makes them even more attractive: estimations show that, e.g., for the single-frequency pumping-based preparation of vacuum–four-exciton states in tetramer or vacuum–three-exciton states in trimer, almost prohibitive laser intensities are needed.

In the current paper we analyze these two aforementioned aspects of the problem. Single- and biharmonic-pulsed laser control of excitons in dimers, trimers, and tetramers of resonantly interacting fluorescent particles are considered, and conditions necessary to prepare different entangled states for them are given with a special emphasis on using a biharmonic pumping. By rescaling, our results can be easily used for any type of particles: quantum dots, dye molecules, or rare-earth dopant ions. To give a fully developed and ready-to-be-used analysis, we illustrate the situation using experimental parameters characteristic for the rare-earth dopant ions, namely, for the three-valent neodymium ions in a cal-

cium fluoride crystal. This is motivated not only by our recent experiments demonstrating the preparation of entangled vacuum and single exciton, as well as vacuum and biexciton states for the pairs of such ions (M centers),[11] but also by the existence of the "naturally designed" tetramers composed by such ions (N centers).[18,23] These M and N centers were under intensive investigation during past years,[24–28] and are investigated nowadays aimed exactly at the realization of the quantum states considered here. The coherent nature of interion interactions for these crystals has been established by us earlier and the characteristic parameters of the decoherence were measured,[27] which enabled us to quantitatively include decoherence into our theoretical analysis.

II. MODEL

Following a widely used approach for this type of problem, resonantly interacting fluorescent particles can be considered as two-level quantum systems (atoms) and treated in a half-integer (pseudo) spin $s=\frac{1}{2}$ formalism. The state with the projection $s_z=-\frac{1}{2}$ describes an atom occupying a ground level while the state $s_z=+\frac{1}{2}$ describes an excited one; for brevity below we will omit the notion "pseudo" and will speak simply about the spin. Let us consider coupled two-level atoms in an external electromagnetic field. Hamiltonian H of this system can be presented as a sum of Hamiltonians corresponding to noninteracting atoms H_i, Hamiltonians of resonant interatomic interaction H_{ij}, and Hamiltonians H_{1i}, describing the interaction of atoms with an external electromagnetic field,

$$H=\sum_i H_i+\sum_{i<j} H_{ij}+\sum_i H_{1i}. \tag{1}$$

We shall assume that all particles are similar and the distances between them are equal, hence

$$H_{ij}=-\hbar V(s_{+i}s_{-j}+s_{-i}s_{+j}). \tag{2}$$

This means that a system formed by equivalent and mutually equidistant fluorescence particles is studied, that is, dimers, trimers (particles occupy the vortices of a right triangle), and tetramers (four particles occupy the vortices of a right tetrahedral).

Under the above assumptions Hamiltonian (1) can be expressed in terms of total-spin operators $\hat{J}_\alpha=\Sigma_i s_{\alpha i}$ ($\alpha=x,y,z$), and for the linearly polarized laser radiation at the frequency ω' and possessing the electric field E, it can be written in the following way:

$$H=H_0+H_{\text{int}}, \tag{3}$$

$$H_0=\hbar\omega_0\hat{J}_z+\hbar V(\hat{J}_z^2-\hat{J}^2), \tag{4}$$

$$H_{\text{int}}=-2d_1E_1\hat{J}_x\cos(\omega' t+\varphi). \tag{5}$$

Here d is the appropriate resonant dipole moment of an atom and $\hbar\omega_0$ is the energy of its excited state. Operators H_0 and $\hat{J}^2,\hat{J}_z$ commute, which means that the well-known angular functions $|J,M\rangle$ (eigenstates of $\hat{J}^2,\hat{J}_z$ operators) are at the same time the eigenstates of Hamiltonian H_0. Hence below these same functions $|J,M\rangle$ will be used to designate the states of the system. For example, for the case of dimers the state $|J=1,J_z=-1\rangle$, or simply $|-1\rangle$, corresponds to the vacuum level, the state $|J=1,J_z=0\rangle\equiv|0\rangle$ corresponds to the single exciton, and so on. Similar notation is used when the density matrix ρ-based description is appropriate: for dimers this means that, say, ρ_{-1-1} describes the population of the vacuum level $|J=1,J_z=-1\rangle\equiv|-1\rangle$, ρ_{00} describes the population of the single-exciton level $|0\rangle$, $\rho_{-1,1}$ describes the coherency between the vacuum level $|-1\rangle$ and the two-exciton level $|1\rangle$, and so on. Correspondingly, wave functions with $J=2$ and J_z ranging from -2 (vacuum) to $+2$ (four exciton) or, for the density-matrix description, indices from -2 to $+2$ and their appropriate combinations are used for the case of tetramer.

It was shown (see, for example, Refs. 1 and 29 and also the Appendix) that in the rotating-frame approximation at the frequency $\omega=\omega'+\Delta\omega$, the resulting Hamiltonian is given by the following equation:

$$\hat{H}_r=\hbar(\omega_0-\omega)\hat{J}_z+\hbar V(\hat{J}_z^2-\hat{J}^2)-d_1E_1\cos(\Delta\omega t-\varphi)\hat{J}_x+d_1E_1\sin(\Delta\omega t-\varphi)J_y. \tag{6}$$

Taking $\Delta\omega=0$, one is able to completely eliminate the time dependence from Hamiltonian; setting also $\varphi=0$ (this just means the choice of the time reference), we obtain

$$\hat{H}_r=\hbar(\omega_0-\omega')\hat{J}_z+\hbar V(\hat{J}_z^2-\hat{J}^2)-d_1E_1\hat{J}_x. \tag{7}$$

For the case of biharmonic pumping light-interaction Hamiltonian is of the more complex form

$$H_{\text{int}}=-2d_1E_1\hat{J}_x\cos(\omega' t+\varphi_1)-2d_2E_2J_x\cos(\omega'' t+\varphi_2) \tag{8}$$

and an explicit dependence on time cannot be eliminated from this Hamiltonian using any transformation to the rotating coordinate frame. The simplest (and usually the most suitable for numerical calculations) form of Hamiltonian can be obtained setting $\omega=\omega'$ and $\varphi_1=0$;

$$\hat{H}_r=\hbar(\omega_0-\omega')\hat{J}_z+\hbar V(\hat{J}_z^2-\hat{J}^2)-d_1E_1J_x-d_2E_2\cos(\Delta\omega t-\varphi_2)\hat{J}_x+d_1E_1\sin(\Delta\omega t-\varphi_2)J_y. \tag{9}$$

Here $\Delta\omega=\omega'-\omega''$. As usual, the quantum dynamics of the system is governed by the master equation $i\hbar\dot{\rho}=[H,\rho]$ with an appropriate Hamiltonian and initial conditions.

To analyze the decoherence processes, we used the following Lindblad-type master equation shown to be appropriate in the Markov random-process approximation for the system at hand:[15,17]

$$i\hbar\frac{\partial\rho}{\partial t}=[H,\rho]-i\hbar\Gamma[J_z[J_z,\rho]]. \tag{10}$$

The calculations presented in the paper were fulfilled using different modifications of Hamiltonian (7) or, when biharmonic pumping is considered, of Hamiltonian (9). To

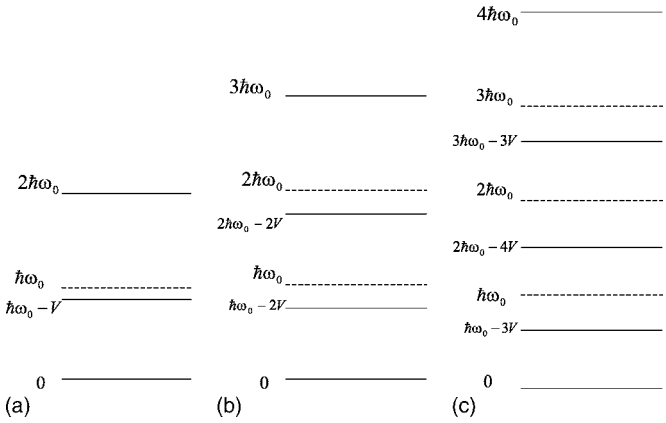

FIG. 1. Energy levels of clusters of two (a), three (b), and four (c) coherently interacting ions.

simulate quantum dynamics, the said differential equations were solved numerically using the MATLAB package. Some of the results were additionally checked and confirmed by computing in the MATHEMATICA package. For clarity, the exponential depopulation of excited states caused by the longitudinal relaxation is not included in the graphs. To distinguish the curves describing the population from those describing entanglement, the latter (to be precise, the absolute values of the nondiagonal elements of the density matrix as a measure of entanglement) were plotted in the lower halves of the figures.

The results of our numerical calculations were confirmed using a specially constructed model: we have showed that an analytical solution for the case of biharmonic pumping of two resonantly interacting particles can be found at appropriate conditions. In the Appendix, we discuss the corresponding results in necessary detail and demonstrate an excellent agreement between numerical and analytical calculations.

III. RESULTS

In the absence of laser irradiation, Hamiltonians (3) and (4) describe rather simple energy structures of the clusters of resonantly interacting fluorescent particles. These structures are shown in Fig. 1. Below we separately discuss the cases of dimers and tetramers. An analysis of the trimer case does not pose any problem (indeed, we have all the necessary results of calculations at our disposal), and we do not present it here, only to save space. Additionally, we would like to underline again the circumstance that while appropriate tetramers (four equivalent fluorescent particles occupied the vortices of a right tetrahedral) do occur in nature (*N* centers of Nd^{3+} ions in different fluoride crystals), to "engineer" an appropriate trimer from, e.g., quantum dots or dye molecules, is not an easy task at all.

The following experimental parameters were used when calculating the numerical results presented below: the decoherence rate at the temperature $T=8$ K was taken as $\Gamma=6.3\ \mathrm{ns}^{-1}$ for dimers (*M* centers) and as $\Gamma=3.5\ \mathrm{ns}^{-1}$ for tetramers;[27] the strength of the interion resonant interaction $V=0.28\ \mathrm{cm}^{-1}$ (Ref. 26) and the resonant-dipole moment of the neodymium ion at the 579.3 nm $^4I_{9/2}$-$^4G_{5/2}$ optical transition $d=8\times10^{-21}$ cgs units[11,26] were used for both cases. Based on the known data on the dependence of the decoherence rate on the temperature,[30] two to three times smaller values of Γ can be anticipated for the temperature of around 4 K and approximately ten times smaller for $T=1$ K. Both these temperature values are quite suitable for experiments and hence the correspondingly modified data was used in our calculations. Moreover, at the temperature $T=4$ K some experiments with *M* and *N* centers in CaF_2 have been already performed.

In our consideration we ignore the Kramers nature of neodymium ions. In practice, this circumstance leads to a more complex character of the interion resonant interaction and, in particular, it is responsible for the additional splitting of the energy levels, cf. Refs. 24 and 26. There are two reasons for ignoring the Kramers nature of the neodymium ions: first, nothing like this exists for the case of quantum dots and dye molecules, and we have in mind a quite general consideration, which could be applied (scaled) for different physical systems. Second, and this is more important, earlier we showed that different energy sublevels, appearing as a result of such a Kramers splitting, can be treated separately with their own values of the measure of the interion interaction *V* and resonant dipole moment *d*.[11] Hence our treatment remains fully applicable with (possible) minor modifications. Currently we are preparing a paper where the full quantum-mechanical consideration of tetramers composed by Kramers neodymium ions will be given.

Having in mind the eventual use of standard broadly available tunable nanosecond lasers in future experiments, we search for such parameters of laser radiation that allow us to achieve the required state during the time of 0.5–5 nanoseconds. To facilitate the rescaling of results, laser intensity is given supposing that the spectral width of the laser line coincides with the homogeneous width of the quantum transition at question. If necessary, the difference between them can be easily taken into account, cf. Ref. 11.

A. Dimers

The energy structure of dimers contains only three optically bright levels [see Fig. 1(a)] and different possibilities of the *single-frequency* control of such a system have been discussed many times theoretically and realized experimentally. The pure single-exciton state $|0\rangle$ and Bell entangled vacuum–single-exciton state $|\psi\rangle_{-10}=(1/\sqrt{2})(|-1\rangle+|0\rangle)$ (Ref. 31) can be prepared by applying an appropriate low-intensity laser pulse at the frequency $\hbar\omega_0-V$. This case was analyzed in detail, e.g., in Ref. 11, and hence will not be considered here.

Another interesting possibility to create a Bell state consists in the preparation of the entangled state of the vacuum $|-1\rangle$ and two-exciton $|1\rangle$ levels: $|\psi\rangle_{-11}=(1/\sqrt{2})(|-1\rangle+|1\rangle)$. One possibility to achieve this, which has been intensively discussed theoretically and realized experimentally, consists in using the single-frequency pumping at the central frequency $\hbar\omega_0$: the biexciton level is then populated via a two-photon absorption process facilitated by the presence of an

intermediate near-resonant single-exciton level shifted only by a rather small value V from an exact resonance. This is illustrated in Fig. 2(a). In Fig. 2(b) we show that the same state can be very efficiently populated by the simultaneous application of two laser pulses: one at the frequency $\hbar\omega_0 - V$ and the other at the frequency $\hbar\omega_0 + V$. Evidently, the state in question is now populated in a stepwise fashion via two single-photon transitions, that is, the single-exciton state is used as an auxiliary intermediate state. This is clearly seen in Fig. 2(b): first, a Bell fully entangled vacuum–single-exciton state $|\psi\rangle_{-10}=(1/\sqrt{2})(|-1\rangle+|0\rangle)$ is achieved (at time $t=1.0$ ns), then a pure single-exciton state is populated ($t=1.9$ ns), and only afterwards subsequent quantum evolution prepares a Bell fully entangled vacuum two-exciton state at $t=3.8$ ns. A comparison of Fig. 2(a) with Fig. 2(b) clearly demonstrates that the quality of prepared states is quite the same while ca. one hundred less-intensive laser pulses are needed to achieve the goal via a biharmonic laser pumping.

By varying the ratio of the laser intensities at different frequencies $I_{\hbar\omega-V}/I_{\hbar\omega+V}$, one can prepare high-quality entangled states of all possible types. This is illustrated in Fig. 2(c): with the ratio $I_{\hbar\omega-V}/I_{\hbar\omega+V}=4$; for different durations of the laser pulse all three possible bipartite Bell states are efficiently prepared: vacuum–single-exciton fully entangled state $|\psi\rangle_{-10}$ is achieved at 1 ns (the value of the corresponding element ρ_{0-1} of the density matrix is 0.49), $|\psi\rangle_{10}$ is achieved at 2.5 ns (with $\rho_{01}=0.45$) and $|\psi\rangle_{1-1}$—at 3.7 ns, $\rho_{1-1}=0.47$. More perfect preparation of any particular Bell state or pure-exciton states can be achieved by further fine tuning of the ratio $I_{\hbar\omega-V}/I_{\hbar\omega+V}$.

The case of equal intensities $I_{\hbar\omega-V}/I_{\hbar\omega+V}=1$ is especially important because just here we have an analytical solution (see the Appendix). Both analytical and numerical results are presented in Fig. 3. As we point out in the Appendix, the analytical solution is valid under condition $2A \ll V$. For the parameters of laser radiation used in Fig. 3(a) (which are exactly those that are necessary from the viewpoint of an experiment), both solutions are virtually undistinguishable. Hence to reveal the validity and limitations of our numerical calculations and analytical model, in Fig. 3(b) we present both solutions for larger laser intensities, where the difference between the two approaches becomes noticeable. (Decoherence effects for the dimers were studied in sufficient detail in our recent paper[11] and we will not reproduce these results here.)

B. Tetramers

Much more reach structure of the energy levels, appearing in the case of tetramers [see Fig. 1(c)], enables us to consider more interesting quantum operations under the system. Earlier, essentially only the possibility to prepare a Greenberger-Horne-Zeilinger state[22] of the vacuum and four exciton $|\psi\rangle_{GHZ}=(1/\sqrt{2})(|-2\rangle+|2\rangle)$ has been analyzed. Although such a state can be created via a single-frequency laser pumping at the central frequency, this implies a four-photon process, which in practice amounts to almost prohibitive laser intensity needed. As we have stated earlier, it seems more practical to consider the preparation and exploration of other entangled states characterized by lower energy.

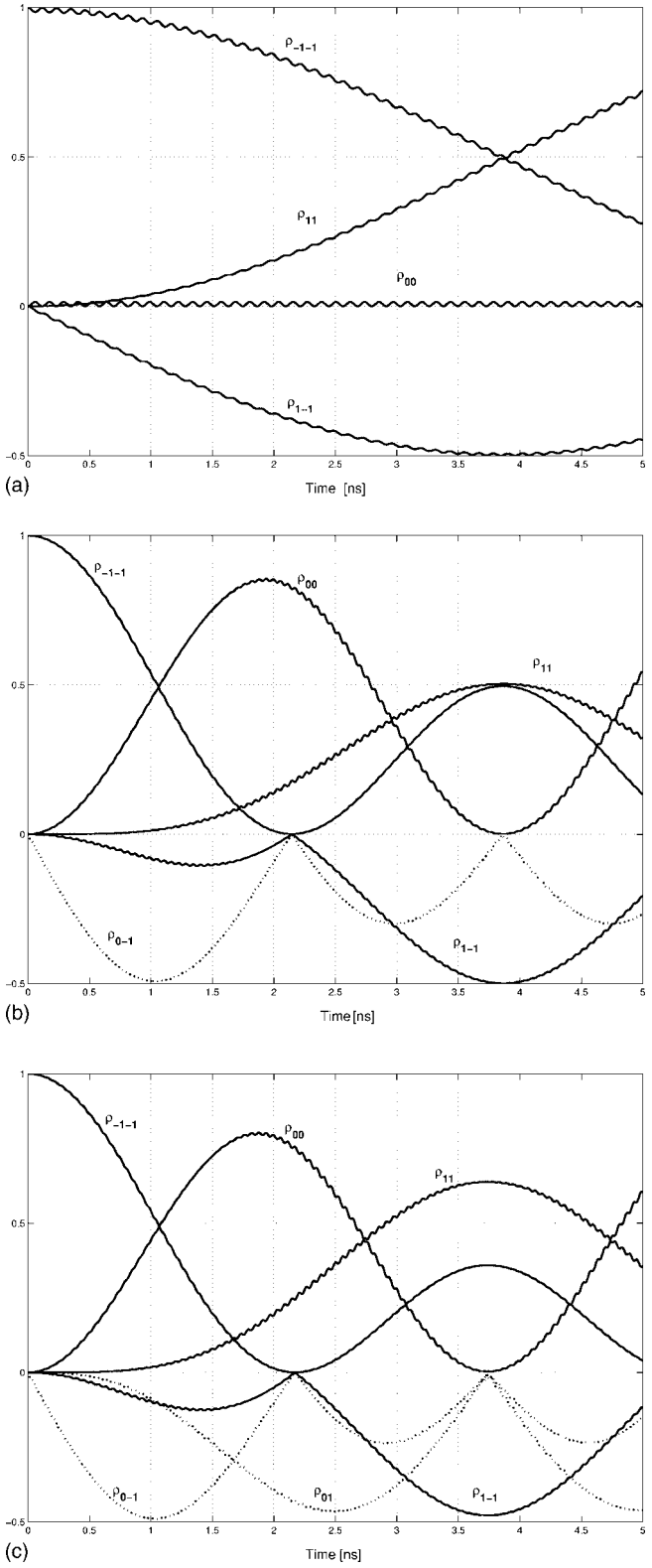

FIG. 2. Effective population of vacuum-biexciton Bell state in dimer for the pumping: (a) at single frequency $\hbar\omega_0$, $I=4.3\times10^7$ W/cm^2; (b) biharmonic at the frequencies $\hbar\omega_0-V$, $I=2.3\times10^6$ W/cm^2, and $\hbar\omega_0+V$, $I=3.9\times10^5$ W/cm^2; (c) biharmonic at the frequencies $\hbar\omega_0-V$, $I=2.3\times10^6$ W/cm^2 and $\hbar\omega_0+V$, $I=5.6\times10^5$ W/cm^2. Other entangled exciton states are also effectively populated using this intensities ratio.

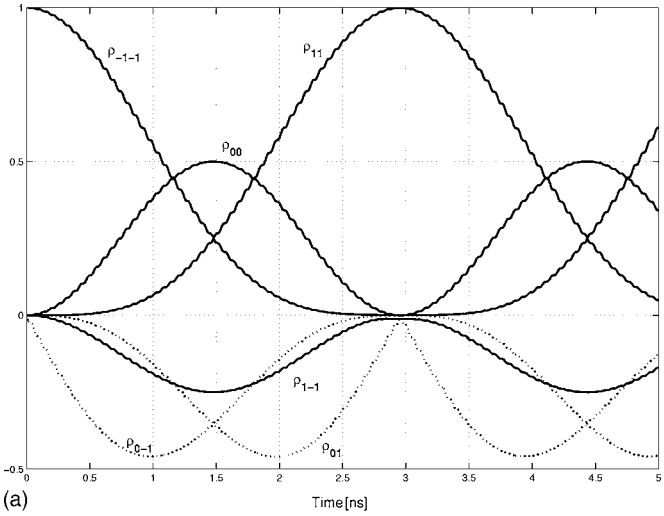

FIG. 3. Population of pure biexciton state in dimer for the case of biharmonic pumping at frequencies ω_0-V and $\omega_0+V$with the same intensity I. Analytical results are obtained with the use of formula (A13) and are shown by a dotted line; numerical results are shown with a solid line. (a) $I=2.3\times10^6$ W/cm^2. The analytical curve coincides with that obtained by numerical calculations and is not visible. (b) $I=6.3\times10^8$ W/cm^2. The difference of two curves becomes noticeable.

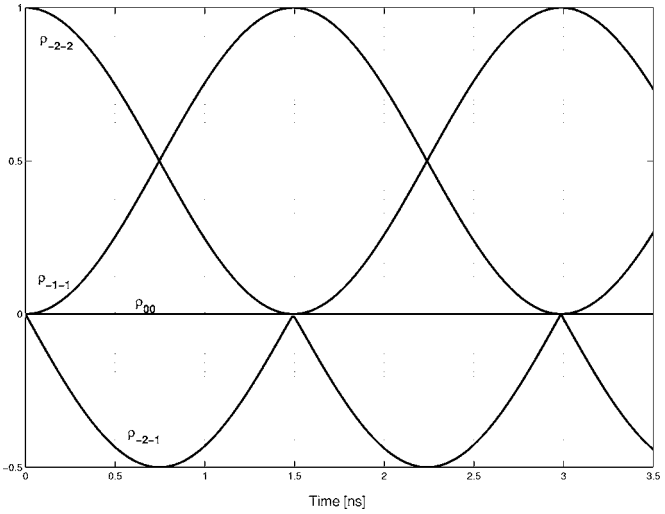

FIG. 4. Effective population of a vacuum–one-exciton Bell state and of a pure one-exciton state in tetramer for the case of single-frequency pumping at $\hbar\omega_0-3V$ with the intensity $I=2.3\times10^6$ W/cm^2.

Let us start our analysis with the preparation of the pure single-exciton state $|-1\rangle$ and the vacuum single-exciton fully entangled state $|\psi\rangle_{-2-1}=(1/\sqrt{2})(|-2\rangle+|-1\rangle)$. This can be done by applying an appropriate laser pulse at the frequency $\hbar\omega_0-3V$. The results of calculations are presented in Fig. 4. This case is almost equivalent to the similar case of the dimer.[11]

A second possibility consists in the preparation of the pure biexciton state $|0\rangle$ and Bell entangled vacuum two-exciton state $|\psi\rangle_{-20}=(1/\sqrt{2})(|-2\rangle+|0\rangle)$. This can be done by virtue of one of two processes: either in a single-frequency fashion by applying laser pulses at the frequency $\hbar\omega_0-2V$, or in a biharmonic fashion by simultaneously applying laser pulses at the frequencies $\hbar\omega_0-3V$ and $\hbar\omega_0-V$. The results of calculations are presented in Fig. 5. Again, the quality of the prepared states is quite similar while a ca. forty-times-smaller intensity is required when the biharmonic pumping is used. This situation is also very similar to the analogous case of the dimer, cf. Fig. 2.

Further, we consider the preparation of the pure three-exciton state and Bell vacuum three-exciton state $|\psi\rangle_{-21}=(1/\sqrt{2})(|-2\rangle+|1\rangle)$. To prepare them in a single-frequency fashion, in principle one could apply a laser pulse at the frequency $\hbar\omega_0-V$. However, as was already stated, this implies a three-photon process and necessitates the usage of almost prohibitively intense pulses. Two other possibilities consist in a combination of one two-photon and one single-photon processes: one can use either pulses at the frequencies $\hbar\omega_0-2V$ and $\hbar\omega_0+V$ [hence the two-exciton level is used as an intermediate one during the excitation process, cf. Fig. 1(c)] or pulses at the frequencies $\hbar\omega_0-3V$ and $\hbar\omega_0$ (the single-exciton level is used as an intermediate one). The results of calculations are shown in Fig. 6.

The pure four-exciton state and Bell vacuum four-exciton state can be prepared via a biharmonic pumping at the frequencies $\hbar\omega_0-2V$ and $\hbar\omega_0+2V$ (a combination of two two-photon processes is needed) with a reasonable fidelity, as attested by Fig. 7.

Other types of entangled states, e.g., a Bell state of two-excitons–three-excitons levels $|\psi\rangle_{01}=(1/\sqrt{2})(|0\rangle+|1\rangle)$, etc., also can be efficiently populated in a biharmonic fashion using the appropriate frequencies and intensities of laser irradiation, and this has been revealed by our calculations. See, for example, the data given in Fig. 6(c), where at the moment $t=3.8$ ns, the matrix element $|\rho_{01}|$ attains the value of 0.45. We analyzed also a few more complicated quantum logic operations under the system, and have shown that they can be successfully performed. To illustrate, in Fig. 8 we show that the system, which initially occupied the fully entangled vacuum three-exciton state $|\psi\rangle_{-21}=(1/\sqrt{2})(|-2\rangle+|1\rangle)$ can be with almost 100% fidelity transferred to the fully entangled single-biexciton state $|\psi\rangle_{-10}=(1/\sqrt{2})(|-1\rangle$

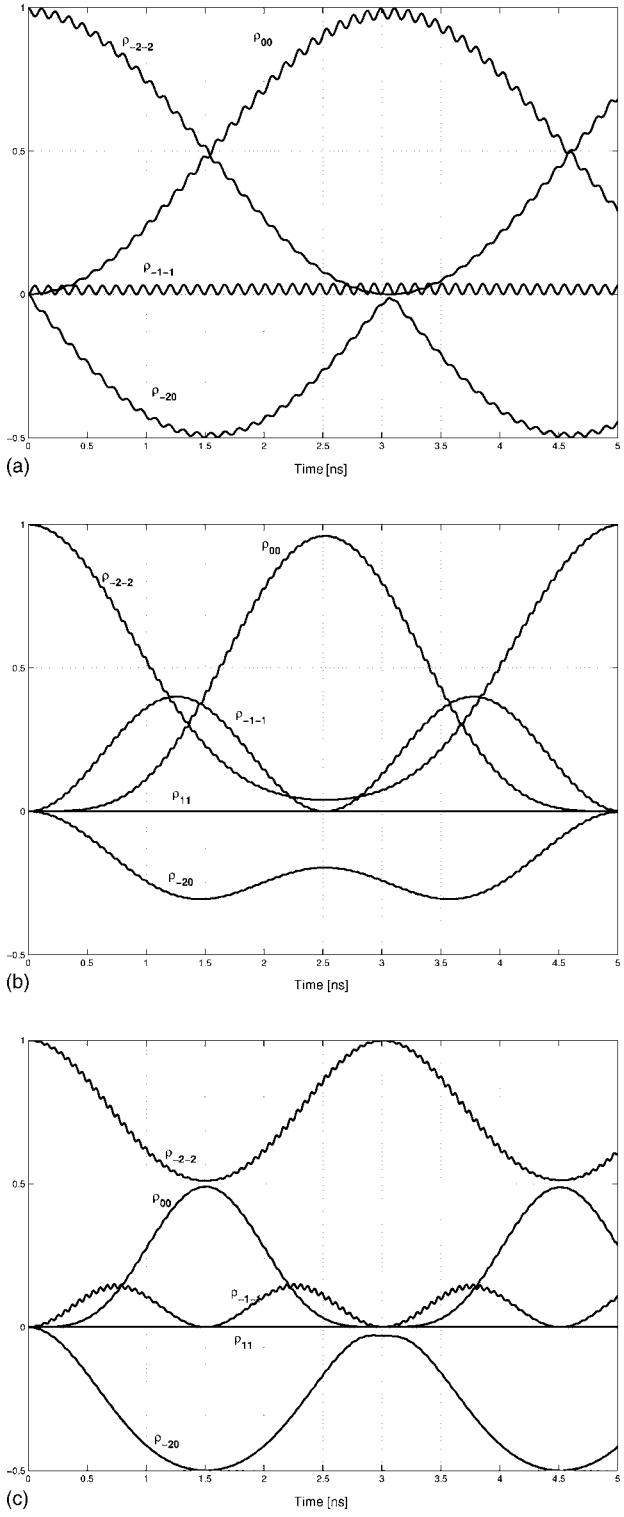

FIG. 5. (a) Effective population of a vacuum-biexciton Bell state and of a pure biexciton state in tetramer for the case of single-frequency pumping at $\hbar\omega_0 - 2V$ with the intensity $I = 4.6 \times 10^7$ W/cm^2. (b) Effective population of a pure biexciton state in tetramer for the case of biharmonic pumping at frequencies $\hbar\omega_0 - 3V$ and $\hbar\omega_0 - V$ with the same intensities $I = 1.3 \times 10^6$ W/cm^2. (c) Effective population of a vacuum-biexciton Bell state in tetramer for the case of biharmonic pumping at the frequencies $\hbar\omega_0 - 3V$, $I = 1.3 \times 10^6$ W/cm^2 and $\hbar\omega_0 - V$, $I = 5.1 \times 10^6$ W/cm^2.

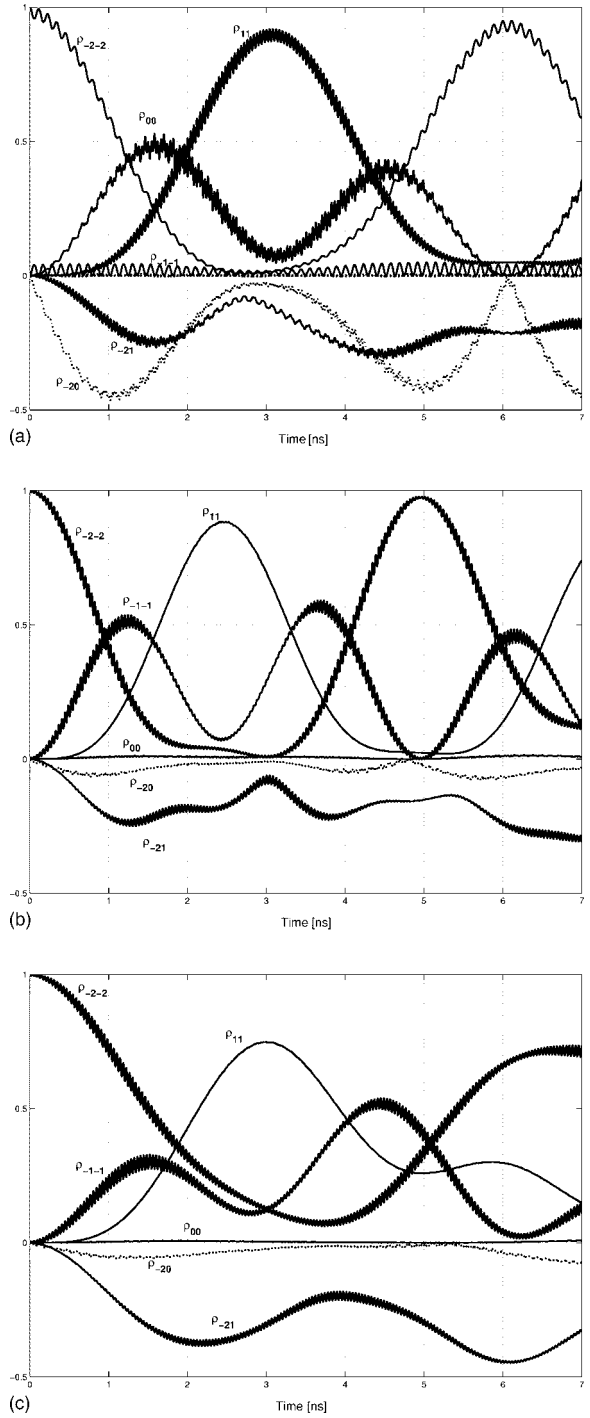

FIG. 6. (a) Effective population of a pure three-exciton state in tetramer for the case of biharmonic pumping at the frequencies $\hbar\omega_0 - 2V$ (two-photon transition, $I = 6.2 \times 10^7$ W/cm^2) and $\hbar\omega_0 + V$ (one-photon transition, $I = 6.9 \times 10^5$ W/cm^2). (b) Effective population of a pure three-exciton state in tetramer for the case of biharmonic pumping at the frequencies $\hbar\omega_0 - 3V$ (one-photon transition, intensity $I = 1.7 \times 10^6$ W/cm^2) and $\hbar\omega_0$ (two-photon transition, $I = 6.2 \times 10^7$ W/cm^2). (c) Effective population of a vacuum–three-exciton Bell state in tetramer for the case of biharmonic pumping at the frequencies $\hbar\omega_0 - 3V$ (one-photon transition, $I = 6.9 \times 10^5$ W/cm^2) and $\hbar\omega_0$ (two-photon transition, $I = 6.2 \times 10^7$ W/cm^2).

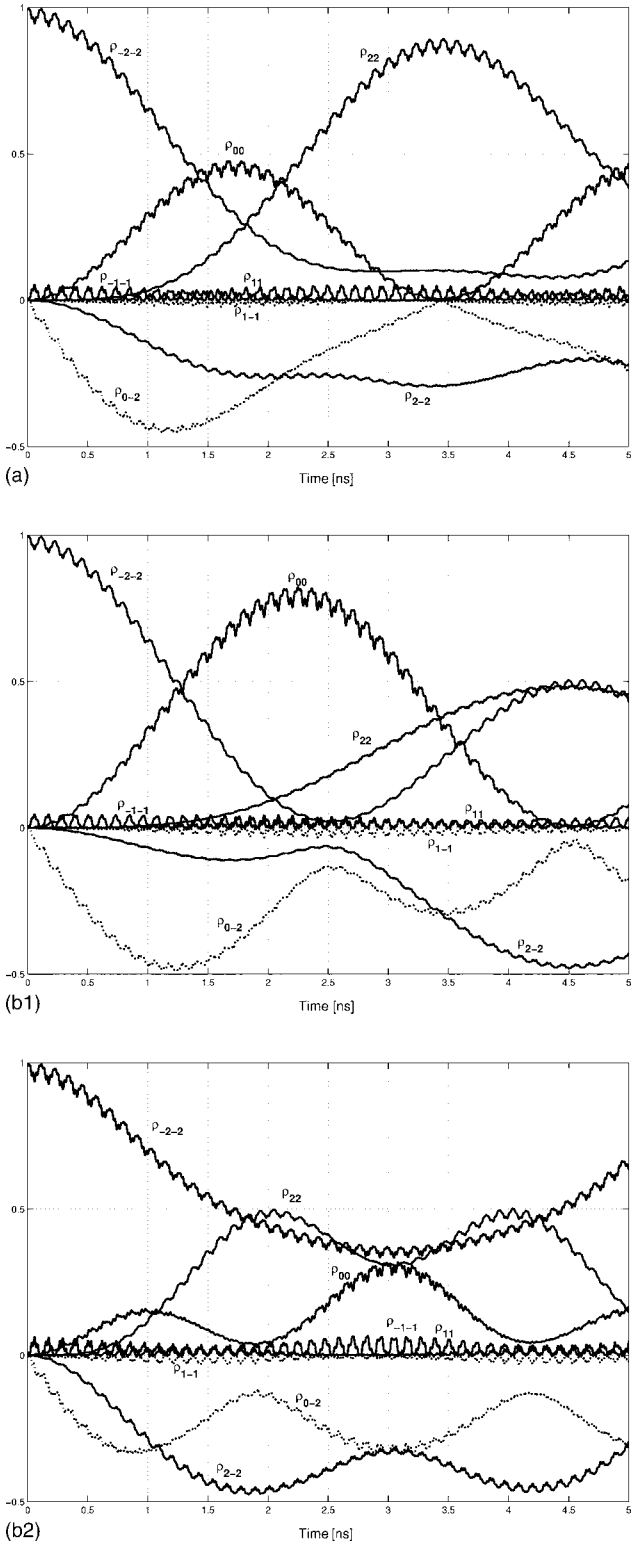

FIG. 7. (a) Effective population of a pure four-exciton state in tetramer for the case of biharmonic pumping at the frequencies $\hbar\omega_0-2V$ and $\hbar\omega_0+2V$ with the same intensity $I=5.6\times10^7$ W/cm^2. (b) Effective population of a vacuum–four-exciton Bell state in tetramer for the case of biharmonic pumping at the frequencies $\hbar\omega_0-2V$ (I_1) and $\hbar\omega_0+2V$ (I_2) with the following intensities: (b1) $I_1=5.6\times10^7$ W/cm^2, $I_2=2.5\times10^7$ W/cm^2; (b2) $I_1=5.6\times10^7$ W/cm^2, $I_2=1.3\times10^8$ W/cm^2.

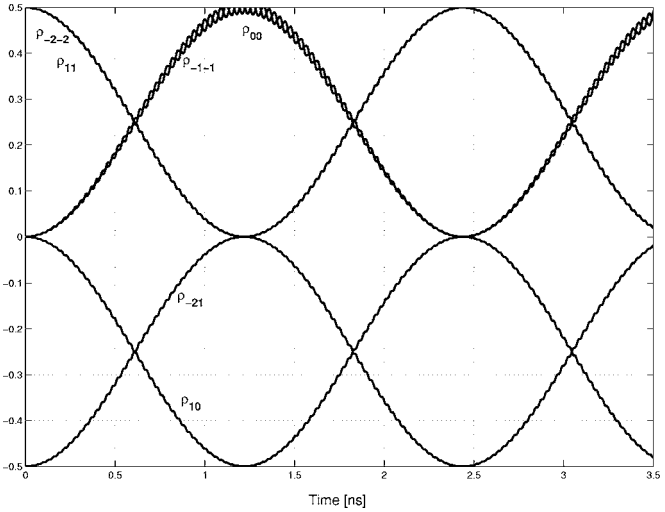

FIG. 8. Complete transfer of a Bell vacuum–three-exciton state $|\psi\rangle_{-21}=(1/\sqrt{2})(|-2\rangle+|1\rangle)$ to a Bell single-biexciton state for the case of biharmonic pumping at the frequencies $\hbar\omega_0-3V$ ($I=3.4\times10^6$ W/cm^2) and $\hbar\omega_0+V$ ($I=2.3\times10^6$ W/cm^2).

$+|0\rangle)$ by the simultaneous action of laser pulses at the frequencies $\hbar\omega_0-3V$ and $\hbar\omega_0+V$. We are not able to present all results
of our calculations here due to the lack of space. To summarize, we can firmly state that these results demonstrate that the excitons in the clusters of resonantly interacting fluorescent particles can be efficiently controlled via a biharmonic laser pumping, and this is not an exaggeration to say that all necessary types of the quantum logic operators (see the discussion of the question what operators are needed, e.g., in Refs. 3 and 12–18) can be realized.

In Fig. 9 we present the results of our investigations of decoherence for the case of tetramers. To illustrate the degree of decoherence we use the normalized linear entropy given by Munro *et al.*[32] with the necessary correction for the tetramer case: $S_L(\rho)=(5/4)\{1-\mathrm{Tr}[\rho^2]\}$. This value ranges from 0 (for pure state) to 1 (for a maximally mixed state). We have plotted the value of S_L along with the absolute values of nondiagonal elements of the density matrix in the lower half of the figures. The same conclusions as for the case of dimers (M centers)[11] can be drawn: although nonnegligible coherency and quantum correlations survive up to the time of a few nanoseconds (especially for the lowest temperatures available), to attain the high fidelity of the quantum operations, usage of tunable lasers with the pulse duration of the order of 0.5–1.5 ns (nowadays these are well available; see, e.g., Ref. 33) is very desirable.

IV. CONCLUSIONS

We have demonstrated that excitons in the clusters of resonantly interacting fluorescent particles can be efficiently prepared and controlled via a biharmonic laser pumping. Although some difficulties still persist, our results indicate that the usage of such biharmonic pumping has important advantages in comparison with the usually analyzed single-

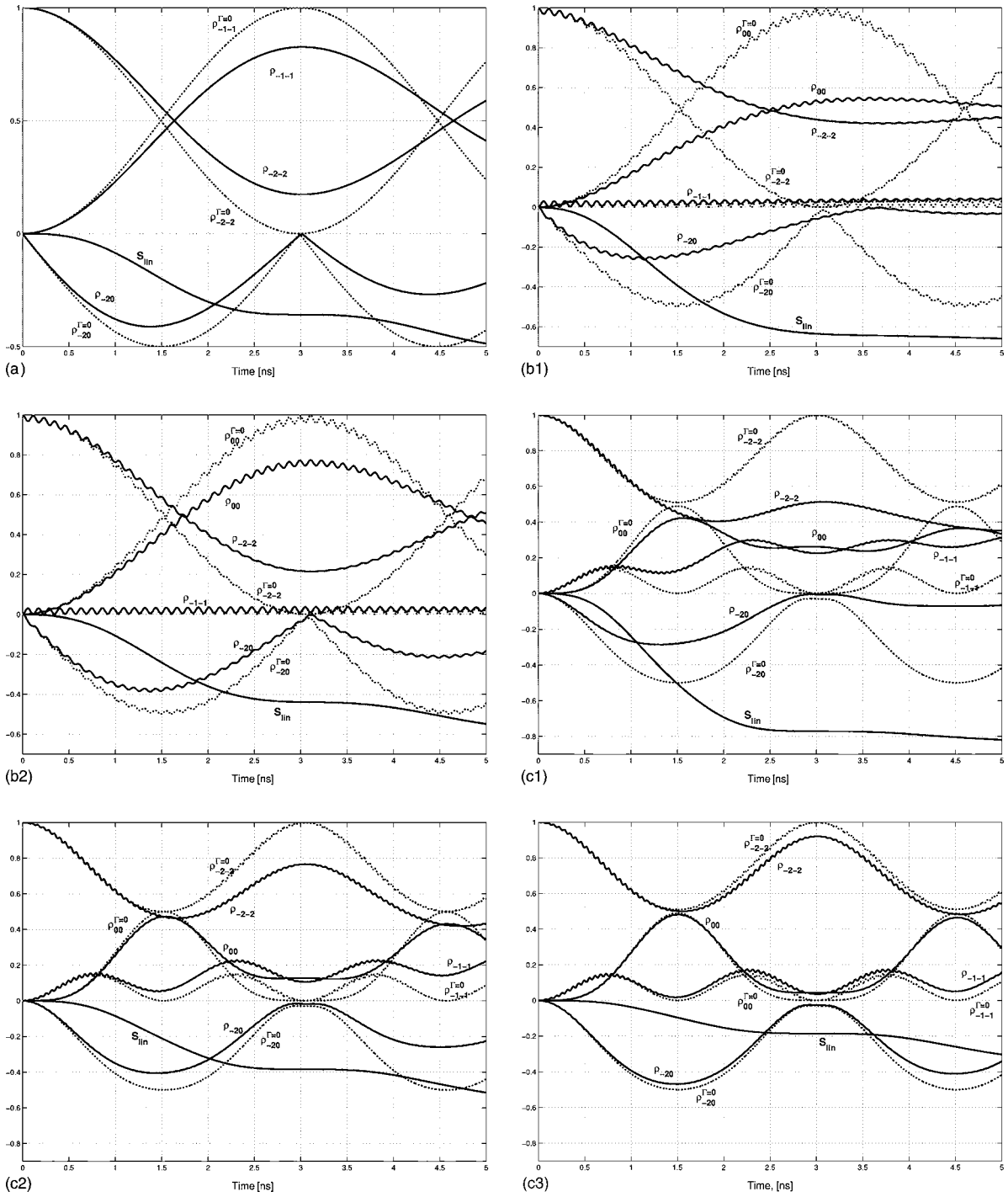

FIG. 9. Quantum dynamics of the tetramers in the presence of decoherence. The quantum dynamics in the absence of decoherence is shown by dotted lines. (a) Evolution of a vacuum–one-exciton Bell state and of a pure one-exciton state for the case of pumping at the single frequency $\hbar\omega_0 - 3V$, $I = 5.6 \times 10^5$ W/cm^2, $\Gamma = 3.5$ ns^{-1}. (b) Evolution of a vacuum-biexciton Bell state and of a pure biexciton state for the case of pumping at the single frequency $\hbar\omega_0 - 2V$, $I = 6.2 \times 10^7$ W/cm^2, decoherence rates: (b1) $\Gamma = 3.5$ ns^{-1} and (b2) $\Gamma = 10.5$ ns^{-1}. (c) Evolution of a vacuum-biexciton Bell state for the case of biharmonic pumping at the frequencies $\hbar\omega_0 - 3V$ ($I = 1.3 \times 10^6$ W/cm^2) and $\hbar\omega_0 - V$ ($I = 5.1 \times 10^6$ W/cm^2), decoherence rates: (c1) $\Gamma = 3.5$ ns^{-1}, (c2) $\Gamma = 10.5$ ns^{-1}, and (c3) $\Gamma = 35$ ns^{-1}.

frequency pumping. Quite realistic values of the laser intensity are needed to realize all types of the quantum logic operations considered here, and indeed we have already demonstrated the practical applicability of exactly such pulses for the case of dimers (M centers of neodymium ions in calcium fluoride crystal).[11] Stepwise biharmonic pumping with the tunable nanosecond lasers is a routine procedure for atoms and molecules,[33] and has been already successfully used for rare-earth ions doped crystals.[34,35]

To conclude, we would like to make the following remark. It is instructive to analyze different states considered in the paper by looking at the localization of excitation en-

ergy on certain ions, which compose the cluster. Let us consider the tetramer and designate a number from 1 to 4 to its constituting ions. Let us also assign the value of 0 to the ion if it is not excited and the value of 1 if it is. In such a notation the vacuum level is simply $|0000\rangle$, the four exciton is $|1111\rangle$, the Bell vacuum four-exciton state is $|\psi\rangle = (1/\sqrt{2})(|0000\rangle + |1111\rangle)$, and so on. (Exactly the latter expression makes the Greenberger-Horne-Zeilinger nature of the Bell vacuum four-exciton state immediately recognizable.) Correspondingly, the vacuum single-exciton fully entangled state is written as

$$|\psi\rangle_{-2-1} = \frac{1}{\sqrt{2}}|0000\rangle + \frac{1}{2\sqrt{2}}(|1000\rangle + |0100\rangle + |0010\rangle + |0001\rangle), \tag{11}$$

the vacuum biexciton fully entangled state is written as

$$|\psi\rangle_{-20} = \frac{1}{\sqrt{2}}|0000\rangle + \frac{1}{2\sqrt{3}}(|1100\rangle + |1010\rangle + |1001\rangle + |0110\rangle + |0101\rangle + |0011\rangle) \tag{12}$$

and so on.

Let us now rewrite the state similar to Eq. (12) in the following form, which explicitly factorizes the quantum state of the first ion as $|0\rangle$ or $|1\rangle$:

$$|\psi\rangle = \frac{1}{\sqrt{7}}|0\rangle(|000\rangle + |110\rangle + |101\rangle + |011\rangle) + \frac{1}{\sqrt{7}}|1\rangle(|100\rangle + |010\rangle + |001\rangle). \tag{13}$$

(We have changed the weights of the vacuum and biexciton states to make the state look more symmetrical.) In such a notation the "maximal connectedness" and/or "persistency of entanglement" of this state[36] become clear: in a general case, the full set of three quantum measurements is needed to "disentangle" it. For example, if the first measurement performed under the ion number one gives "0," it projects the system into the "highly entangled" state $(1/\sqrt{2})(|000\rangle + |110\rangle + |101\rangle + |011\rangle)$. The subsequent measurement performed under the ion number two (suppose we once again obtain "0") projects it into still entangled state $(1/\sqrt{2})(|00\rangle + |11\rangle)$, and hence a final additional measurement is needed to achieve the full disentanglement. Such states with a large connectedness, often named simply "cluster states," nowadays start to be considered as an important resource for the quantum computing,[36–40] and this definitely increases an interest in different low-energy exciton entangled states considered in the paper. Of course, readout of the quantum state in such a system is a complicated process, which deserves a special discussion. However, we believe that the exploitation of laser excitation at dipole-allowed *f-d* transitions could be really useful here, as it was briefly discussed in our earlier work.[19]

Note that although this type of decomposition of exciton states with the explicit indication of the localization of an excitation energy on certain fluorescent ions is applied starting from apparently the first work in the field (Ref. 1), we believe that only now, in the context of our recent proposal to use low-temperature scanning probe microscopy to handle relatively long-lived fluorescent rare-earth ions,[11,18–20] such a decomposition becomes really substantial. The proposed approach does allow us to separate spatially the ions comprising the cluster and hence to perform the measurements under a certain specific ion, which makes this consideration meaningful.

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APPENDIX

Following the common procedure, let us transform light-interaction Hamiltonian $H_{\text{int}} = -2dEJ_x \cos(\omega' t + \varphi')$ to the coordinate frame rotating with the frequency ω; hereafter we will use a tilde to distinguish the operators and variables acting in such a frame. To calculate the corresponding Hamiltonian, that is, to find the value of $\tilde{H}_{\text{int}} = e^{i(\omega t+\varphi)J_z} H_{\text{int}} e^{-i(\omega t+\varphi)J_z}$, one should apply the known relation

$$e^{\lambda J_z} J_{\pm} e^{-\lambda J_z} = e^{\pm\lambda} J_{\pm}. \tag{A1}$$

Setting $\lambda = i(\omega t + \varphi)$, we obtain

$$\begin{aligned}\tilde{H}_{\text{int}} &= -2dE[J_x \cos(\omega t + \varphi) - J_y \sin(\omega t + \varphi)]\cos(\omega' t + \varphi') \\ &= -dEJ_x\{\cos[(\omega + \omega')t + \varphi + \varphi'] \\ &\quad + \cos[(\omega - \omega')t + \varphi - \varphi']\} + dEJ_y \\ &\quad \times\{\sin[(\omega + \omega')t + \varphi + \varphi'] + \sin[(\omega - \omega')t + \varphi - \varphi']\}.\end{aligned}$$

When the condition $\omega' \approx \omega$ holds, rapidly oscillating terms can be neglected (the rotating-wave approximation, which is valid when $\hbar\omega \gg |d_1 E_1|, |d_2 E_2|$), and this expression shortens to $\tilde{H}_{\text{int}} = -dEJ_x \cos[(\omega - \omega')t + \varphi - \varphi'] + dEJ_y \sin[(\omega - \omega')t + \varphi - \varphi']$. For the case of exact coincidence of the frequencies $\omega' = \omega$ and of the phases $\varphi = \varphi'$, one obtains famous $\tilde{H}_{\text{int}} = -dEJ_x$ thus completely eliminating an explicit dependence on time.

Now let us consider the particular case of two resonantly interacting fluorescent particles subject to the biharmonic pumping (we imply that $V \ll \omega$) $H_{\text{int}} = -\{2d_1 E_1 \cos[(\omega - V)t + \varphi_1] + 2d_2 E_2 \cos[(\omega + V)t + \varphi_2]\}J_x$ and perform the transformation to the frame rotating at the central frequency ω with phase φ. This gives

$$\begin{aligned}\tilde{H}_{\text{int}} &= -d_1 E_1[\cos(Vt + \varphi - \varphi_1)J_x - \sin(Vt + \varphi - \varphi_1)J_y] \\ &\quad - d_2 E_2[\cos(Vt - \varphi + \varphi_2)J_x + \sin(Vt - \varphi + \varphi_2)J_y].\end{aligned} \tag{A2}$$

Now let us restrict ourselves with the case of equal intensities $d_1E_1=d_2E_2$. One can use the remaining freedom of the selection of the phase φ to set $\varphi=(\varphi_1+\varphi_2)/2$. As a result, the terms containing J_y are canceled, and the Hamiltonian governing the behavior of coherently interacting particles takes the form

$$\tilde{H} = \hbar\, VJ_z^2 + 2\hbar\, A\cos(Vt+\varphi_{21})J_x, \tag{A3}$$

where we denoted $A=-d_1E_1/\hbar=-d_2E_2/\hbar$ and $\varphi_{21}=(\varphi_2-\varphi_1)/2$.

After the changing of our standard basis $|1\rangle$, $|0\rangle$, and $|-1\rangle$, corresponding to the definite values of the total pseudospin projection J_z, to the basis $|f_1\rangle=(1/\sqrt{2})[|1\rangle-|-1\rangle]$, $|f_2\rangle=(1/\sqrt{2})[|1\rangle+|-1\rangle]$, $|f_3\rangle=|0\rangle$, Hamiltonian (A3) is transformed into the following form:

$$\tilde{H}_f/\hbar = \begin{pmatrix} V/2 & 0 & 0 \\ 0 & V/2 & 2A\cos(Vt) \\ 0 & 2A\cos(Vt) & -V/2 \end{pmatrix}$$

$$= \begin{pmatrix} V/2 & 0 & 0 \\ 0 & & VS_z \\ 0 & & \end{pmatrix} + \begin{pmatrix} 0 & 0 & 0 \\ 0 & & 4A\cos(Vt)S_x \\ 0 & & \end{pmatrix}. \tag{A4}$$

Here S_α $(\alpha=x,y,z)$ are components of $\frac{1}{2}$-spin operator $\boldsymbol{S}$, we set $\varphi_{21}=0$ and explicitly write this Hamiltonian as a sum of diagonal and nondiagonal parts. Its quasidiagonal structure enables us to simplify it further by introducing matrix $\hat{Z}$,

$$\hat{Z} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1/2 & 0 \\ 0 & 0 & -1/2 \end{pmatrix} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & & \\ 0 & & S_z \end{pmatrix}, \tag{A5}$$

and performing the transformation $\tilde{H}_f'=P\tilde{H}_fP^+$, where $P=e^{iV\hat{Z}t}$, $P^+=e^{-iV\hat{Z}t}$. This quasirotation exploits the matrix $\hat{Z}$ instead of J_z in the manner completely similar to that given above when discussing the transformation to the frame rotating at the frequency ω. Taking into account a rule for multiplying the quasidiagonal matrices and commutation rules for S_α operators, we have

$$\tilde{H}_f'/\hbar = \begin{pmatrix} -V/2 & 0 & 0 \\ 0 & & 2AS_x \\ 0 & & \end{pmatrix} + \begin{pmatrix} 0 & 0 & 0 \\ 0 & & A(S_+e^{2iVt}+S_-e^{-2iVt}) \\ 0 & & \end{pmatrix}. \tag{A6}$$

Neglecting the terms with the frequencies $2Vt$ (second rotating-wave approximation, which is valid when the condition $2A \ll V$ holds), we discard the second term in Eq. (A6) thus obtaining a time-independent Hamiltonian entering the master equation $\dot{\tilde{\rho}}'=-(i/\hbar)[\tilde{H}_f',\tilde{\rho}']$ for which a solution is easy to obtain. We transform this solution back to the coordination frame used before the last transformation under the matrix $\hat{Z}$. Taking into account that $\tilde{\rho}'(0)=\tilde{\rho}(0)=\rho(0)$, introducing the abbreviation $\theta=At$ and matrix R to shorten the final expression

$$R = e^{-iV\hat{Z}t}e^{-i\tilde{H}_\varphi t/\hbar} = \begin{pmatrix} e^{-iVt/2} & 0 & 0 \\ 0 & e^{-iVt/2}\cos\theta & -ie^{-iVt/2}\sin\theta \\ 0 & -ie^{iVt/2}\sin\theta & e^{iVt/2}\cos\theta \end{pmatrix}, \tag{A7}$$

the following solution in the rotating frame, basis f_1, f_2, f_3, is obtained:

$$\tilde{\rho} = R\rho(0)R^+. \tag{A8}$$

Returning back to the basis $|M\rangle$ we obtain the following explicit expression for matrix R:

$$R = \begin{pmatrix} & |1\rangle & |0\rangle & |-1\rangle \\ \langle 1| & (\cos\theta+1)/2 & (-i/\sqrt{2})\sin\theta & (\cos\theta-1)/2 \\ \langle 0| & (-i/\sqrt{2})e^{iVt}\sin\theta & e^{iVt}\cos\theta & (-i/\sqrt{2})e^{iVt}\sin\theta \\ \langle -1| & (\cos\theta-1)/2 & (-i/\sqrt{2})\sin\theta & (\cos\theta+1)/2 \end{pmatrix}. \tag{A9}$$

If an initial state of the system is a pure one, that is, $\rho(0)$ can be represented as $\rho(0)=|\psi(0)\rangle\langle\psi(0)|$, then $\tilde{\rho}(t)$ can be rewritten in the form

$$\tilde{\rho}(t) = |\tilde{\psi}(t)\rangle\langle\tilde{\psi}(t)| \tag{A10}$$

with

$$|\tilde{\psi}(t)\rangle = R|\psi(0)\rangle. \tag{A11}$$

For our initial conditions $\rho(0)=|-1\rangle\langle -1|$, we have for $|\psi(t)\rangle$ in the rotating frame the simple expression

$$|\tilde{\psi}(t)\rangle = R|-1\rangle = \frac{1}{2}(\cos\theta - 1)|1\rangle - \frac{i}{\sqrt{2}}e^{iVt}\sin\theta|0\rangle + \frac{1}{2}(\cos\theta + 1)|-1\rangle. \tag{A12}$$

Finally, an explicit form of result (A10) for initial conditions $\rho(0)=|-1\rangle\langle -1|$ is given by the expression

$$\tilde{\rho}(t) = \begin{pmatrix} & |1\rangle & |0\rangle & |-1\rangle \\ \langle 1| & \frac{1}{4}(\cos\theta-1)^2 & \frac{i}{2\sqrt{2}}e^{-iVt}\sin\theta(\cos\theta-1) & -\frac{1}{4}\sin^2\theta \\ \langle 0| & \frac{-i}{2\sqrt{2}}e^{iVt}\sin\theta(\cos\theta-1) & \frac{1}{2}\sin^2\theta & \frac{-i}{2\sqrt{2}}e^{iVt}\sin\theta(1+\cos\theta) \\ \langle -1| & -\frac{1}{4}\sin^2\theta & \frac{i}{2\sqrt{2}}e^{-iVt}\sin\theta(1+\cos\theta) & \frac{1}{4}(1+\cos\theta)^2. \end{pmatrix}. \tag{A13}$$

A comparison of the results given by Eq. (A13) and those of numerical calculations is presented in Fig. 3 and discussed in Sec. III A.

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