Multipulse laser spectroscopy of $\overline{p}\,\mathrm{He}^+$: Measurement and control of the metastable state populations

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Numerical study of the kinetics in metastable antiprotonic helium molecules driven by a sequence of laser π pulses resonant to the adjacent transitions is carried out. The phenomena taking place are shown to be useful for checking the mechanisms of the depopulation of high-n states, for measuring the populations of the individual energy levels, and for slowing down the decay by means of π-pulse-induced inversion. [S1050-2947(97)02204-X]

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I. INTRODUCTION

The recently discovered metastable states of antiprotons in helium [1] are of significant interest as a means of accumulating antimatter in neutral atoms. This is important for further use of antimatter in new energy sources as well as in fundamental experiments on CPT-invariance testing, gravitation interaction of antiparticles, etc. [2]. Besides that, antiprotonic atoms, as well as other hadronic and muonic atomic and molecular systems, are themselves a very interesting object for the improvement and testing of theoretical and computational tools of the few-body quantum physics.

That is why the antiprotonic atoms became a subject of intense theoretical and experimental studies, resulting in a growing number of publications and international meetings. A comprehensive study of long-lived antiprotons in helium has been carried out using the superior antiproton beam from LEAR (Low Energy Antiproton Ring) at CERN [3–9].

The phenomenon of delayed annihilation of antiprotons in helium may be well interpreted in terms of metastable states of large (n,l) of a neutral exotic system $\overline{p}\,\mathrm{He}^+$, as suggested by Condo [10] and studied theoretically by Russell [11] more than 20 years ago. Recently, new theoretical treatments of this system have been developed. Yamazaki and Ohtsuki [12] calculated the energy levels and their radiative lifetimes using the configuration mixing method including various orbitals of the antiproton and electron, established the approximate selection rules for radiative transitions, and estimated the Auger transition rates.

Another theoretical approach, molecular approach, was developed by Shimamura [13]. In this model the He nucleus and the antiproton are regarded as the two centers of an exotic molecule, to which one electron is coupled adiabatically. The results obtained in the energy levels and the radiative transition rates are nearly the same as those of the atomic approach. Thus $\overline{p}\,\mathrm{He}^+$ possesses a dual character in itself both as an atom and as a molecule.

Delayed annihilation time spectra (DATS) systematically studied in various phases of helium and also in helium media with foreign atoms or molecules [3,5] have provided important information on the formation and decay of the metastable molecules, but it is a cumulative, integral, and macroscopic effect and difficult to interpret in terms of individual states involved. ''Differential and microscopic'' information can be provided by laser resonance spectroscopy [14] in which transitions stimulated by a pulsed laser produce a spike in DATS at the laser timing. Such a resonance spike was observed in 1993 by Morita *et al.* [6,7] on the 597 nm (39,35)-(38,34) transition. In 1994 they found an additional transition, (37,34)-(36,33) [8]. Hayano *et al.* [7,8] deduced the lifetimes and populations from the measurements of the time dependence of the first laser spike and the recovery time detected by the second successive laser tuned at the same wavelength. Their results are in excellent agreement with the theoretically calculated radiative rates [13,15]. Simultaneous excitation of adjacent transitions $3\rightarrow2\rightarrow1$ by using two lasers has also been proven experimentally [16].

Recently a theoretical treatment using the molecular-based large configuration variational method has been carried out by Korobov [17]. Calculations of the transition energies using this method stimulated new experiments [18]. Nearly perfect agreement between the theory and experiment up to the level of 50 ppm was demonstrated.

Thus the present state of quantum mechanical treatment of $\overline{p}\,\mathrm{He}^+$ is sufficient to provide one with reliable estimates of energies, transition moments, and radiation lifetimes. However, there exist no rigorous quantum mechanical calculations of the initial populations of metastable states in $\overline{p}\,\mathrm{He}^+$. Quasiclassical estimations of these populations [20] based on the theoretical approach of [12,15,21] demonstrate satisfactory agreement with DATS experiments [14], provided that the levels with $n\geqslant41$ are populated negligibly small due to efficient collisional quenching [21,22]. According to [22] the lifetime of the states with $n\geqslant41$ is less than 1 ns. The knowledge of the initial population distribution is important for the control of metastable atom decay kinetics

and for the experimental testing of decay mechanisms of higher levels ($n \geq 41$).

Our numerical estimations show that the total DATS is not critically sensitive to the variation of the initial population distribution. Based on the results of numerical simulations we propose here a coherent laser multipulse technique for control and measurement of the kinetics of metastable states in $\overline{p}\mathrm{He}^+$. The term ''coherent'' means that each pulse in the sequence is short enough compared with the atomic relaxation times so that optical transient phenomena [23] are essential. In particular, we propose a method of direct measurement of the populations of the metastable states at an arbitrary instant of time. Compared to the papers cited above this technique is shown to provide a more selective action on the individual transitions and, as a result, more detailed information on the population distribution.

II. SLOWING DOWN THE DECAY OF THE METASTABLE $\overline{p}\mathrm{He}^+$ BY π-PULSE-INDUCED POPULATION INVERSION

Recent experimental and theoretical studies of the metastable states of $\overline{p}\mathrm{He}^+$ reveal some typical features of the optical transition scheme. Namely, the optical transitions take place between the metastable states including a finite number of energy levels, say $(n,l) \rightarrow (n-1,l-1)$. Each cascade is limited by the nonstable (annihilation) part of the energy spectrum from the bottom and by the states which are efficiently depopulated by external factor (most probably, collisions with He atoms) from the top. The dominant mechanism of decay of each state is the radiative transition to the neighboring lower energy level of the same chain. The initial populations of the lower states are usually greater than those of the upper ones (or, at least, there is no monotonic decrease of population down by the cascade). The lowest metastable state decays straight into a short-lived state, which results in fast annihilation of $\overline{p}$.

As an example we consider the cascade of transitions between the states with $v = n-l-1 = 2$ (Fig. 1) which has been a subject of intense experimental and theoretical studies [8,13] and is known to include the four metastable states.

As is known, a π pulse resonant to a certain transition in a multilevel system having relatively slow relaxation inverts the population of the two coupled levels, leaving the other ones practically unaffected, provided that all transition frequencies are substantially different from each other.

For the level diagram shown in Fig. 1 the separations between the transition frequencies are known to be much greater than the transition linewidth and the typical width of the laser π-pulse spectrum, so that the selectivity of the action of each pulse on a given transition is guaranteed.

To increase the total lifetime, one should prepare the system in the state with the maximal inversion of populations by means of a properly chosen squence of π pulses. The order of π pulses depends upon the initial populations. For example, according to Ohtsuki's calculations [20], the latter decrease monotonically from lower to upper levels. In this case the sequence of π pulse should be $(2 \rightarrow 3)$, $(3 \rightarrow 4)$, $(4 \rightarrow 5)$, $(2 \rightarrow 3)$, $(3 \rightarrow 4)$, $(2 \rightarrow 3)$. For other initial population distributions both the number and the order of pulses may be different.

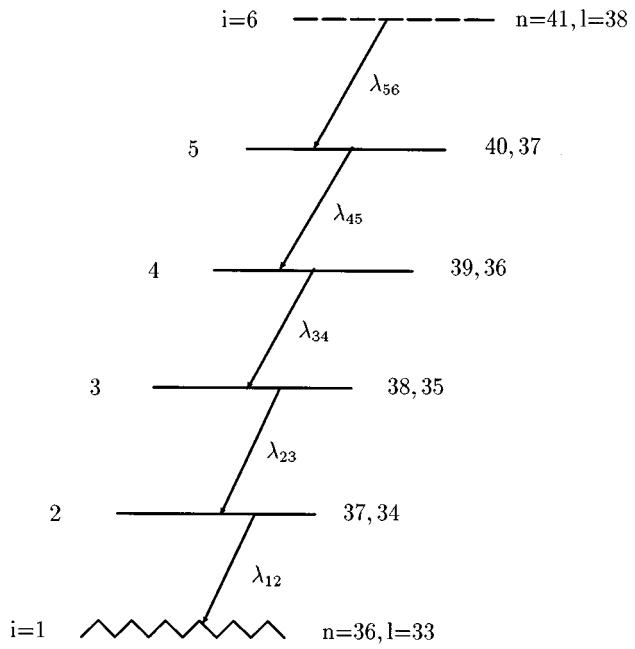

FIG. 1. Part of the energy-level diagram of $\overline{p}\mathrm{He}^+$. Solid lines show the metastable states, zigzag line denotes the short-lived annihilation state, dashed line shows the states depopulated by collisions.

The prolongation of the mean lifetime of the system is thus achieved due to the simple fact that most of the molecules populate the top levels of the scheme and it takes them some longer time to reach the ''annihilation bottom'' via the longer chain of spontaneous radiation transitions. The additional contribution to the effect comes from the slight growth of the radiative decay time from lower to upper states.

The numerical modeling of the coherent laser excitation and decay of the system was done by solving the complete set of density matrix equations, describing the interaction of a multilevel system with multifrequency pulsed radiation:

$$\dot{\hat{\rho}} = -\frac{i}{\hbar}[\hat{H}\hat{\rho}] - \frac{1}{2}(\hat{\Gamma}\hat{\rho} + \hat{\rho}\hat{\Gamma}) + \hat{\Lambda},$$

where $\hat{\rho}$ is the density matrix operator with the matrix elements $\rho_{ik} = \langle i|\hat{\rho}|k\rangle$, $i,k = 1, \ldots, 6$ numerating the levels,

$$\hat{H} = \begin{pmatrix} \hbar\omega_1 & V_{12}(t) & 0 & 0 & 0 & 0 \\ V_{12}(t) & \hbar\omega_2 & V_{23}(t) & 0 & 0 & 0 \\ 0 & V_{23}(t) & \hbar\omega_3 & V_{34}(t) & 0 & 0 \\ 0 & 0 & V_{34}(t) & \hbar\omega_4 & V_{45}(t) & 0 \\ 0 & 0 & 0 & V_{45}(t) & \hbar\omega_5 & V_{56}(t) \\ 0 & 0 & 0 & 0 & V_{56}(t) & \hbar\omega_6 \end{pmatrix},$$

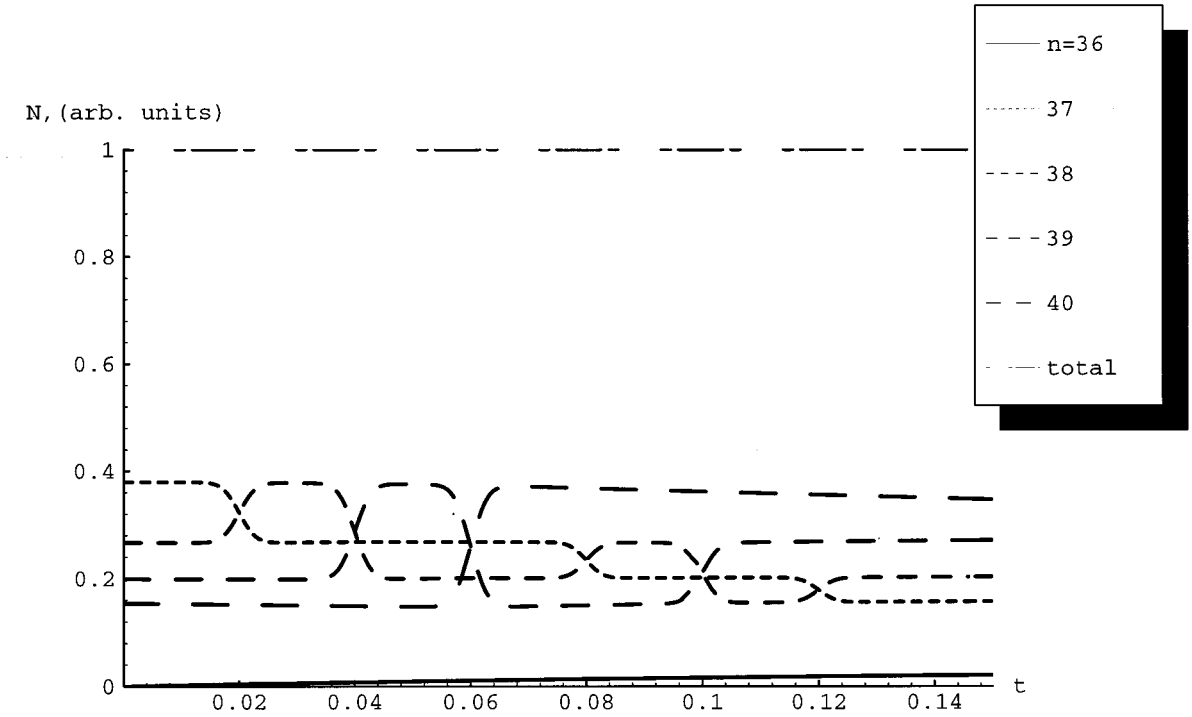

FIG. 2. Inversion of energy-state populations by a sequence of laser π pulses. Here and below the time is expressed in microseconds and the populations are normalized so that the total initial population in the cascade is 1.

$$\hat{\Gamma}=\begin{pmatrix} \gamma_1 & 0 & 0 & 0 & 0 & 0 \\ 0 & \gamma_2 & 0 & 0 & 0 & 0 \\ 0 & 0 & \gamma_3 & 0 & 0 & 0 \\ 0 & 0 & 0 & \gamma_4 & 0 & 0 \\ 0 & 0 & 0 & 0 & \gamma_5 & 0 \\ 0 & 0 & 0 & 0 & 0 & \Gamma_6 \end{pmatrix},$$

$\hbar\omega_i$ is the energy of the ith level, γ_i is the relaxation rate of the ith level, $\Gamma_6=\gamma_6+\gamma_{\rm col}$, and $\gamma_{\rm col}$ is the collisional quenching rate of level 6.

$$\hat{\Lambda}=\begin{pmatrix} \gamma_2\rho_{22} & 0 & 0 & 0 & 0 & 0 \\ 0 & \gamma_3\rho_{33} & 0 & 0 & 0 & 0 \\ 0 & 0 & \gamma_4\rho_{44} & 0 & 0 & 0 \\ 0 & 0 & 0 & \gamma_5\rho_{55} & 0 & 0 \\ 0 & 0 & 0 & 0 & \gamma_6\rho_{66} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix},$$

$\hat{\rho}$ is normalized as $\Sigma_{i=1}{}^{6}\rho_{ii}=1$,

$$V_{ij}(t)=-\sum_m (A_m/2\tau_p\sqrt{\pi})\exp\{-[(t-t_m)/\tau_p]^2 - i[(\omega_i-\omega_j-\nu_m)t]\}+\text{c.c.},$$

A_m is the area of the mth pulse, and ν_m is the pulse carrier frequency. We will suppose below that all pulses are resonant to the corresponding transitions with zero detuning.

In our model the relaxation of the diagonal elements ρ_{kk}, $k=2,\ldots,5$ is caused only by the spontaneous radiation transitions to the next lower level. The collisions are not taken into account in γ_i, so there is no ''elastic'' contribution to the relaxation of coherences ρ_{km}. It is also assumed that states other, than those shown in Fig. 1 are not involved in the relaxation processes. This assumption is based on the fact [9,13] that the appropriate spontaneous transition rates are at least one order smaller than those taken into account. To avoid complicated coherent phenomena the pulses are separated by a short delay from each other, so each of them acts separately on the appropriate transition.

Figure 2 shows the inversion of energy state populations by a sequence of π pulses. One can see sequential interchange of populations between the levels 2 and 3, 3 and 4, etc. The pulse width and amplitude are chosen to satisfy the π-pulse area condition, providing complete inversion of population. For the typical values of laser pulse parameters (pulse duration $\tau_p\simeq 1$ ns, reduced dipole moment $\wp\simeq 1.5\times 10^{-30}$ A s m [19], and the laser beam radius $w\simeq 1$ mm) the necessary pulse power is about 10^2 W, which may be easily achieved using the commercial laser systems.[1]

The whole sequence of pulses is much shorter in time than the typical decay time, so the laser action may be considered as two different scale processes: (i) ''instant'' preparation of inverted system; (ii) free decay of this system. The

[1]Although the antiproton beam radius in LEAR experiments [3–9] could be as small as 1 mm, the $\bar{p}$ stopping region after passing through the window and the helium medium itself was subject to multiple scattering. Computer simulations showed [24] that it is about 10 mm diameter. In this case the transverse profile of the laser beam is, of course, essential. However, the transverse effects in the laser beams are beyond the scope of this study.

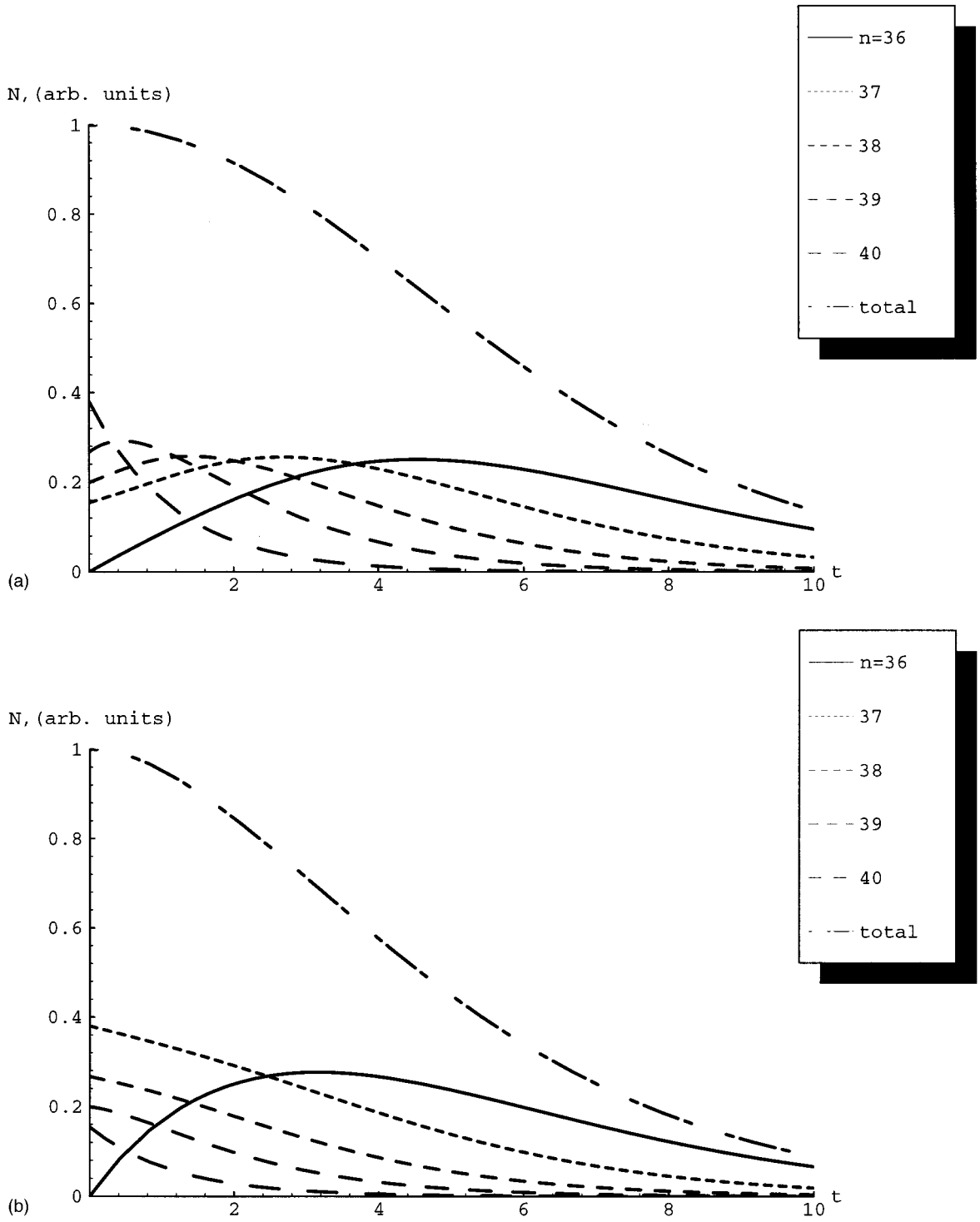

FIG. 3. Large-scale kinetics of decay of individual metastable states previously prepared by the π-pulse sequence (a) compared with that without laser preparation (b).

second part of the temporal evolution may be considered as a spontaneous decay with inverted population distribution which can be simulated using ordinary rate equations for the level populations. In principle, the code used allows one to deal with pulses of arbitrary areas, chirped or detuned pulses, and, if desired, account for coherent effects of simultaneous action of the same pulse on two or more parallel neighboring cascades.

In Fig. 3 the large-scale kinetics of decay of individual metastable states previously prepared by the π-pulse sequence (a) is compared with that without laser preparation (b). The nonmonotonic temporal dependence of populations of levels 2–5 [Fig. 3(a)] is determined by the transition to these states from the upper ones. These transitions compete with the ordinary spontaneous decay of these states.

Figure 4 demonstrates the total population of all metastable levels of the cascade versus time. One can see that the laser-pulse preparation yields an increase of about 20% in

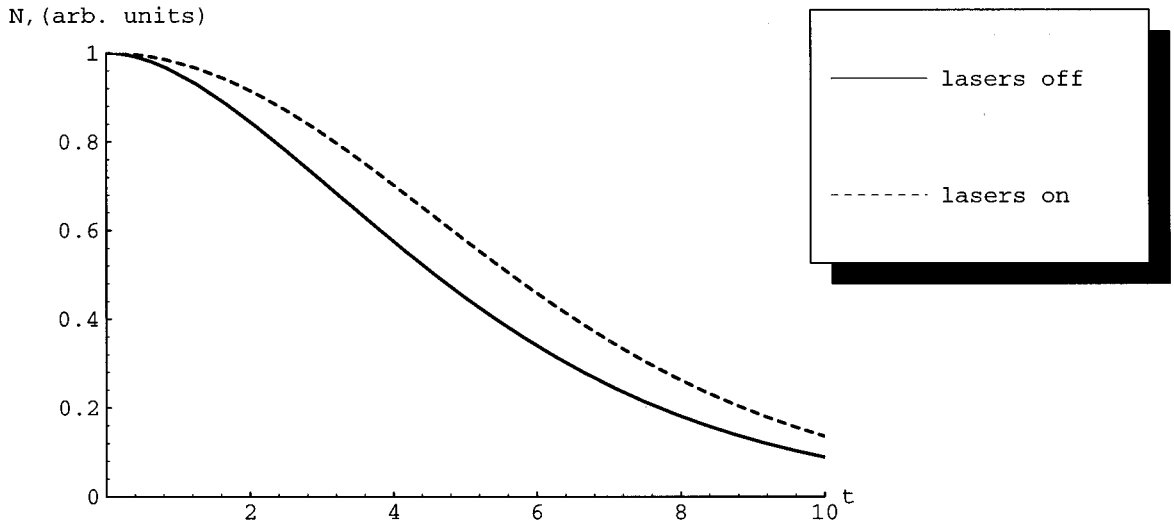

FIG. 4. Total population of all metastable levels of the cascade versus time.

the total number of metastable molecules throughout the decay process. Thus the proposed technique may give a substantial increase of the total number of metastable molecules as well as of the mean lifetime.

Pulses of arbitrary area will, as a rule, produce smaller effect. For example, long pulses or cw radiation will produce only saturation, i.e., equalizing the initially different populations, but they cannot invert the transition. This is the principal idea of this paper as compared with other studies of laser excitation of $\overline{p}\,\mathrm{He}^+$.

III. CHECKING THE MECHANISM OF THE DEPOPULATION OF THE STATES WITH $N \geqslant 41$

The interpretation of DATS experiments [14] implies the anomalously low population of high states ($n \geqslant 41$). The mechanisms of this are not clear within the existing theory of the isolated $\overline{p}\,\mathrm{He}^+$ formation. Ohtsuki's estimations show that there is nothing extraordinary about the initial population of these levels compared with those with $n \leqslant 40$. The most realistic explanation [21,22] of this fact is the fast ($\tau \simeq 1$ ns) quenching due to collisions of the molecule with He atoms. There are no direct experimental data on the rate and channels of the depopulation of the $n \geqslant 41$ states, as well as no evidence in favor of the idea that they are not initially populated at all.

We will show that the multipulse excitation technique allows one to enforce the population of these states and monitor the decay that follows. The efficiency of this procedure is maximal when the population of the neighboring ($n = 40$) level is previously increased as much as possible by means of the inversion sequence.

Suppose, for example, that the state $n = 41$ has a very short lifetime due to the fast collisional quenching resulting in the transition of the moleclue to the states not shown in Fig. 1. Figure 5 shows large-scale decay kinetics after the π-pulse sequence, increasing the population of level 5 ($n = 40$), followed by a π pulse, transmitting the accumulated particles to state 6 ($n = 41$). Thus if the kinetic model supposed in this calculation is valid, then the fast decay of this state will cause a step in the total particle number, followed by the common decay picture.

If the decay of the $n = 41$ state occurs in a usual manner, i.e., via the transitions to the $n = 40$ state, no step in the total population curve takes place, and the decay kinetics is qualitatively the same as discussed in the preceding section. Thus by comparing the experimental results with numerical curves obtained for different decay models one can choose among them.

IV. MEASURING THE POPULATIONS OF INDIVIDUAL STATES

Primary distribution of populations of metastable states is the dominant factor affecting the laser-induced kinetics in $\overline{p}\,\mathrm{He}^+$, discussed in Sec. I. Significant work has been done in experimental study of population dynamics in $\overline{p}\,\mathrm{He}^+$. From the measurement of the time dependence of the first laser spike and the recovery time detected by the second laser pulse tuned to the same transition the initial populations of three levels involved in the $v = 3$ cascade were deduced assuming a certain kinetic model and using the best-fit procedure [7]. It is desirable to have a means of direct measurement of the population distribution at any instant of time. We will show that the multipulse coherent excitation technique using nanosecond pulses separated by nanosecond delays (i.e., practically instantaneous compared to the decay time) provides such an opportunity.

The idea is rather simple. We have already mentioned that a π pulse causes complete inversion of the resonant transition, i.e., the final population of the lower energy state becomes equal to the initial population of the upper energy state and vice versa. Hence the π pulse resonant to the transition $2 \rightarrow 1$ between the lowest metastable state 2 and the neighboring short-lived state 1 enforces the fast annihilation of all antiprotons populating state 2. This results in a spike of the annihilation output measured in a way similar to [6–8]. The spike amplitude is proportional to the population of state 2 at the moment of the pulse action.

If we replace the single pulse $2 \rightarrow 1$ with a sequence of

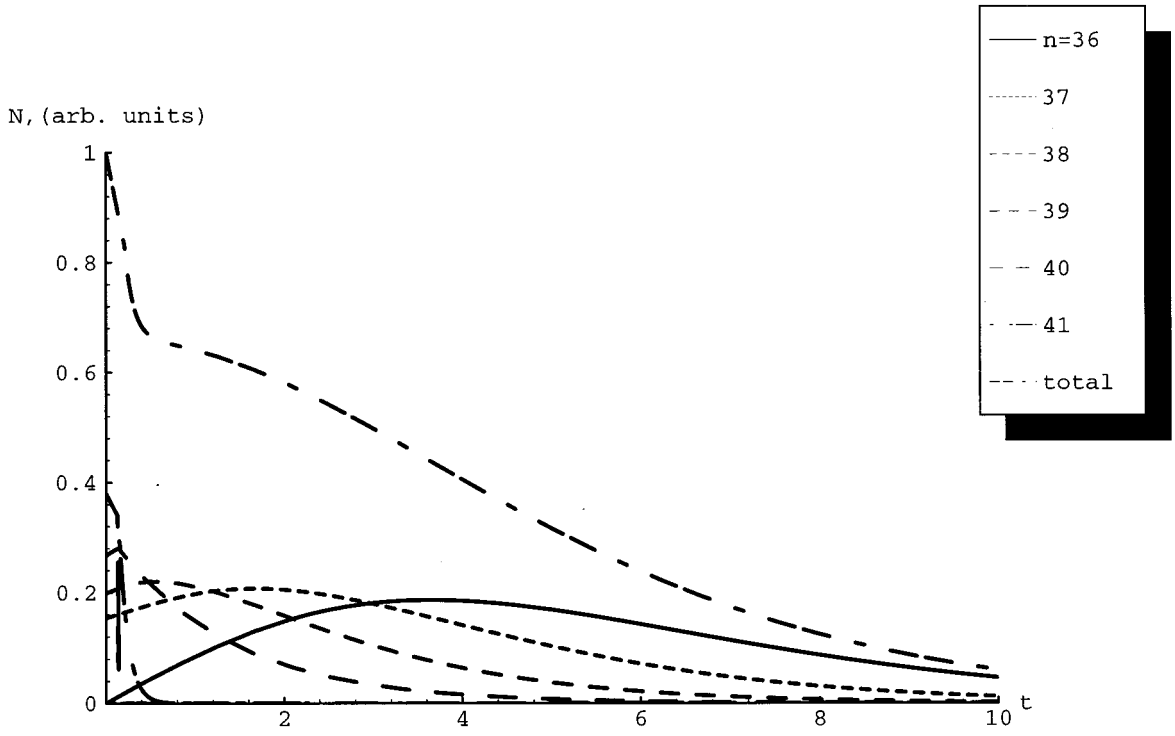

FIG. 5. Large-scale decay kinetics after the π-pulse sequence, increasing the population of level 5 ($n=40$), followed by a π pulse, transmitting the accumulated particles to the collision-quenched state 6 ($n=41$).

three pulses $2\rightarrow1$, $3\rightarrow2$, $2\rightarrow1$ then all molecules which have initially been in states 2 and 3 appear to be shaken down to state 1 and finally cause an annihilation peak proportional to the total initial population of states 2 and 3.

The linear dependence of the annihilation peak upon the populations is due to the fact that the typical π-pulse duration as well as the delays between the pulses are of the order of 10^{-9} s, i.e., the laser action on the system is practically instantaneous compared to the spontaneous decay lifetime (the number of annihilation events is counted during the time of nearly 10 ns).

Performing similar experiments with sequences of pulses resonant to various transitions in the cascade one can observe a series of annihilation peaks, the ratios of their amplitudes giving directly the relative population of individual levels at the moment of the laser action.

The experimental realization of this technique seems to meet no major problems since there exist commercial tunable lasers in the frequency region considered which can provide pulses of necessary length and of power even higher than required.

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