Effective excitation method of a three-level medium in a selective photoionization

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A principle for the efficient excitation of a three-level medium in a selective photoionization method is examined. It is shown that with special detunings for a given set of the laser-atom interaction the third-level population can be maximized. The expression of the optimal detunings is derived near the two-photon resonance region. It is shown that if the optimal detuning method is applied to three-level systems of Yb and Ca, the first-level populations are inverted to their corresponding third levels by about 100% and 86%, respectively. Application of this method to a Doppler-broadened medium is discussed and it is found that the optimal detuning method with counterpropagating waves will be very effective for selective ionization of wanted atoms.

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I. INTRODUCTION

Selective photoionization by a multiphoton process has been of interest in laser spectroscopic fields. As is well known, differences in absorption spectra of isotopes are the fundamental factors in the phenomenon, and the process of photoionization consists of selective excitation of a particular isotope and photoionization from the upper excited level. However, the efficiency and the selectivity of the process should be high for practical application. Although up-to-date laser systems are well developed in the realization of such processes, the appropriate conditions to achieve high efficiency and selectivity usually cannot be known directly without an optimization process. Hence investigations for the optimization process of laser action upon matter are considered to be very important. Such an investigation can be performed from the theoretical point of view. In this paper, we will suggest a principle for optimizing an excitation process of three-level media which enables us to enhance the efficiency of photoionization of wanted atoms in the media, as well as to obtain high selectivity.

Since a selective photoionization is achieved through the process of selective excitations followed by successive ionization from the upper excited level, it can be realized in two ways which have different excitation mechanisms. The first way corresponds to an usual method such as a dynamic photoionization of the medium, where all the laser pulses are simultaneously coupled with the transition levels. This process can be optimized by controlling the ratio of the parameters of the medium and the laser fields. However, our previous numerical investigation showed that the first way could not be effective [1]. In the second way, the medium is first in the situation that the wanted isotopes are fully populated at the third levels, but other isotopes are empty there, and the photoionization radiation pulse is subsequently switched on. In this case the ionization efficiency can be high since the photoionization pulse interacts with all the atoms to be ionized. The above second way of ionization was suggested by Diels [2], who provided only some results of two-level systems. The method, besides a high ionization efficiency, can enhance the selectivity of isotopes.

We applied the Diels idea to a three-level system, and investigated the behavior of a three-level medium by varying the laser frequencies in a region near two-photon resonances, in which the Bloch equation was numerically solved. Particularly in the three-level excitation scheme compared with the two-level one, the ionization coefficient can be enhanced by varying the laser frequencies from the resonant transition ones. Resulting from such a study, it was found that there can be detunings which cause the population of the third level to be maximized for a given set of the medium-field interactions. The detunings were found to be near two-photon resonances and to depend on the energy density and the duration of the laser pulses as well as the dipole moments of the first and the second transition. Here, we will examine the principle of optimal detunings and suggest their analytic formulas in the case of a stationary electromagnetic field, before applying the optimal-detuning method to a real atomic medium.

II. DYNAMICS OF THE THREE-LEVEL ATOM

For the investigation of population dynamics of a three-level atomic system as shown in Fig. 1, we use a theoretical method which is based on the density-matrix formalism. The dynamics of the medium is described by a Liouville equation such that

$$i\hbar\left[\frac{\partial}{\partial t}+\hat{\Gamma}\right]\hat{\rho}=[\hat{H}_0+\hat{V},\hat{\rho}] \ , \tag{1}$$

where $\hat{\rho}$ is the density operator, $\hat{\Gamma}$ the operator of the system relaxation, $\hat{H}_0$ the Hamiltonian of the unperturbed

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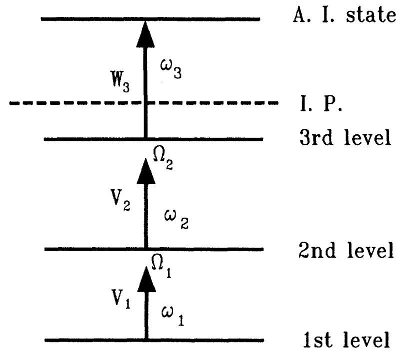

FIG. 1. The photoionization scheme. V_1 and V_2 are Rabi frequencies. Ω_1 and Ω_2 are the detunings of laser frequencies. W_3 is the ionization loss rate. ω_1, ω_2, ω_3 are the laser frequencies.

system, and $\hat{V}$ the operator describing the interaction with the electromagnetic fields. Since we have considered the interaction in a dipole approximation, the operator $\hat{V}$ is given by $\hat{V}=\hat{d}\hat{E}/\hbar$ where the operator $\hat{d}$ corresponds to the dipole moment of the three-level system. We assume that the resonant laser field is a superposition of three linearly polarized plane waves. The eigenfunctions for the density-matrix elements can be written in the resonant approximation. Even though a rigorous description of the three-step photoionization scheme requires a density matrix of 4×4 dimensions, the description can be simplified if we keep in mind that a photoionization process from the third bound level can be incoherently represented. It can be assumed in this case that the laser radiation producing photoionization causes the three-level medium to be only attenuated with a rate of $W_3=\sigma_3 I_3/\hbar\omega_3$, where I_3 is the photoionization light intensity, $\hbar\omega_3$ the photon energy, and σ_3 the photoionization cross section. This assumption allows the density-matrix formalism to be represented in a simple form of 3×3 dimensions, as the photoionization process can be dealt with as one mechanism of system relaxation and it can be described by a conventional kinetic equation of balance.

The Bloch equation describing the interaction for the Doppler-broadened three-level medium with a monochromatic laser field such as

$$\mathbf{E}_j(t)=\mathbf{e}_z \varepsilon_j(t)\exp[i(\omega_j t-\phi_j)]+\text{ c.c.}$$

can be derived without any difficulty [3]. Here, $\mathbf{e}_z$, $\varepsilon_j(t)$, ω_j, and ϕ_j are the unit vector of polarization, the amplitude, the frequency, and the phase of the jth laser field, respectively. That is, the set of equations is given by

$$\frac{\partial}{\partial t}\rho_{11}=-2\xi_1 V_1+A_{21}\rho_{22}\ , \tag{2}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{T_2}\right]\rho_{22}=2(\xi_1 V_1-\xi_2 V_2)+A_{32}\rho_{33}\ , \tag{3}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{T_3}+W_3\right]\rho_{33}=2\xi_2 V_2\ , \tag{4}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{2T_2}\right]u_1=\Omega_1\xi_1-V_2\xi_3\ , \tag{5}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{2T_2}\right]\xi_1=\Omega_1 u_1+V_1(\rho_{11}-\rho_{22})+V_2 u_3\ , \tag{6}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{2}\left[\frac{1}{T_2}+\frac{1}{T_3}+W_3\right]\right]u_2=-\Omega_2\xi_2+V_1\xi_3\ , \tag{7}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{2}\left[\frac{1}{T_2}+\frac{1}{T_3}+W_3\right]\right]\xi_2 =\Omega_2 u_2+V_2(\rho_{22}-\rho_{33})-V_1 u_3\ , \tag{8}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{2}\left[\frac{1}{T_3}+W_3\right]\right]u_3 =-(\Omega_1+\Omega_2)\xi_3+V_1\xi_2-V_2\xi_1\ , \tag{9}$$

$$\left[\frac{\partial}{\partial t}+\frac{1}{2}\left[\frac{1}{T_3}+W_3\right]\right]\xi_3=(\Omega_1+\Omega_2)u_3-V_1 u_2+V_2 u_1\ , \tag{10}$$

$$\frac{\partial}{\partial t}n_+=W_3 n_3\ , \tag{11}$$

$$n_j=\int D(v_y)\rho_{ij}dv_y \quad \text{for } j=1,\ 2,\text{ and } 3\ , \tag{12}$$

$$\Omega_j=\left[\omega_{j+1,j}\left[1-\frac{v_y}{c}\right]-\omega_j\right] \quad \text{for } j=1 \text{ and } 2\ , \tag{13}$$

$$V_j=\frac{d_j\varepsilon_j(t)}{\hbar} \quad \text{for } j=1 \text{ and } 2\ . \tag{14}$$

Here, $u_{1,2,3}$ and $\xi_{1,2,3}$ are the in-phase and 90°-out-of-phase components of the medium polarization, respectively. n_j is the jth-level population, and n_+ is the ion concentration. The Doppler broadening is described by the velocity distribution function $D(v_y)$. d_1 and d_2 are the dipole transition matrices of the first and the second transition, respectively. ω_{21} and ω_{32} are the resonant frequencies of the first and the second transitions, respectively. A_{ij} is the Einstein coefficient of the transitions, c is the light velocity, and $\hbar$ is the Planck constant.

Let us consider the behavior of a three-level atom in a stationary electromagnetic field without any relaxation. Here, we are concerned with only a two-photon resonance region. We get a solution of Eqs. (2)–(14) for such an exact and sharp two-photon resonance as $\Omega=\Omega_1=-\Omega_2$ with $V_1=V_2\ll\Omega$. The solution can be found as follows:

$$\rho_{33}(t)=\frac{4V_1^2V_2^2}{(V_1^2+V_2^2)^2}\sin^2\left[\left[\frac{V_1^2+V_2^2}{8\Omega}\right]t\right]\ . \tag{15}$$

From Eq. (15), we can see that a similar behavior as that of a two-level atom is revealed in the case of three-level atoms, where the corresponding Rabi frequency is $(V_1^2+V_2^2)/4\Omega$.

Considering the behavior of ρ_{33} in the (Ω_1,Ω_2) coordinates along a two-photon resonance line, we can see that the amplitude of ρ_{33} becomes 1 under the condition of V_1 equal to V_2. Hence it is clear that at any intensity and duration of laser pulses the three-level system can be controlled to get a maximum population by varying the two-photon detuning (Ω). In such a case of $T_0(V_1^2+V_2^2)/4\Omega$ being equal to π, the three-level system can be fully inverted. Here, T_0 is half the duration time of a stationary pulse. But in a real situation we cannot always satisfy the requirement that V_1 is equal to V_2. However, here we will show that there are detunings which invert the three-level medium, even when V_1 is not equal to V_2.

Although the system expressed by Eqs. (2)–(14) has an analytical solution on the condition mentioned below [3], it will be very complicated for thorough analysis. So we will approximate the system so that the solution can be analyzed. On the condition of nearly two-photon resonance such that $\Omega=\Omega_1\approx-\Omega_2$, if $\Omega^2\gg V_1^2, V_2^2$, Eqs. (2)–(14) can be written as

$$\frac{\partial}{\partial t}\rho_{11}=-2\xi_1 V_1 , \tag{16}$$

$$\frac{\partial}{\partial t}\rho_{22}=2(\xi_1 V_1-\xi_2 V_2) , \tag{17}$$

$$\frac{\partial}{\partial t}\rho_{33}=2\xi_2 V_2 , \tag{18}$$

$$0=-\Omega_1\xi_1-V_2\xi_3 , \tag{19}$$

$$0=\Omega_1 u_1+V_1(\rho_{11}-\rho_{22})+V_2 u_3 , \tag{20}$$

$$0=-\Omega_2\xi_2+V_1\xi_3 , \tag{21}$$

$$0=\Omega_2 u_2+V_2(\rho_{22}-\rho_{33})-V_1 u_3 , \tag{22}$$

$$\frac{\partial}{\partial t}u_3=-(\Omega_1+\Omega_2)\xi_3+V_1\xi_2-V_2\xi_1 , \tag{23}$$

$$\frac{\partial}{\partial t}\xi_3=(\Omega_1+\Omega_2)u_3-V_1 u_2+V_2 u_1 . \tag{24}$$

In these equations we neglect derivatives such as $\partial u_1/\partial t$, $\partial\xi_1/\partial t$, $\partial u_2/\partial t$, and $\partial\xi_2/\partial t$ because they are much less than $\Omega_1 u_1, \Omega_1\xi_2, \Omega_2 u_2$, and $\Omega_2\xi_2$. That is, their values are of the order of $1/\Omega$. Then the system permits a clear analytical solution for a stationary electromagnetic field, where V_1 and V_2 are constant. The result can be used for a detailed investigation of the excitation dynamics.

The expression for $\rho_{33}(t)$ is given by

$$\rho_{33}(t)=-\frac{V_1^2V_2^2}{4\Omega_1\Omega_2\alpha^2}\sin^2\frac{\alpha t}{2} , \tag{25}$$

where α is

$$\alpha^2=\left[\Omega_1+\Omega_2-\frac{V_1^2}{4\Omega_2}-\frac{V_2^2}{4\Omega_1}\right]^2 +\frac{V_1^2V_2^2}{4}\left[\frac{1}{\Omega_1^2}+\frac{1}{\Omega_1\Omega_2}+\frac{1}{\Omega_2^2}\right] . \tag{26}$$

In the case of a two-photon resonance such that $\Omega_1=-\Omega_2=\Omega$ it can be shown that the solution is exactly the same as Eq. (15). Hence we are sure that the approximation condition on the system is appropriate.

We consider such a condition that $\rho_{33}(t)$ is maximum. We can see from Eqs. (25) and (26) that if the values of V_1 and V_2 are determined the detunings Ω_1 and Ω_2 should satisfy the condition that $\alpha T_0=\pi$ and simultaneously $\Omega_1\Omega_2\alpha^2$ is minimum. Such a particular solution can be given by the following two equations. That is,

$$\Omega_1+\Omega_2-\frac{V_1^2}{4\Omega_2}-\frac{V_2^2}{4\Omega_1}=0 , \tag{27}$$

$$\frac{V_1^2V_2^2}{4}\left[\frac{1}{\Omega_1^2}+\frac{1}{\Omega_1\Omega_2}+\frac{1}{\Omega_2^2}\right]=\left[\frac{\pi}{T_0}\right]^2 . \tag{28}$$

A simpler expression for the optimal detunings can be obtained if we use new variables in the (Ω_1,Ω_2) plane. That is, rotation of the coordinate axes by 45° transforms the variables for the detunings as follows:

$$\Omega_1=\frac{(\Delta_2+\Delta_1)}{\sqrt{2}} , \tag{29}$$

$$\Omega_2=\frac{(\Delta_2-\Delta_1)}{\sqrt{2}} , \tag{30}$$

where Δ_1 and Δ_2 are the parameters characterizing the detunings at and away from the two-photon resonance, respectively. Since we are concerned with a detuning region near the two-photon resonance, we can neglect Δ_2^2 compared with Δ_1^2. Then the expression for ρ_{33} becomes

$$\rho_{33}=\frac{\gamma^2\sin^2\alpha t/2}{(\Delta_2-\beta)^2+\gamma^2} , \tag{31}$$

where

$$\alpha^2=\frac{[4\Delta_2\Delta_1^2+V_1^2(\Delta_2+\Delta_1)+V_2^2(\Delta_2-\Delta_1)]^2}{8\Delta_1^4} +\frac{1}{2}\left[\frac{V_1V_2}{\Delta_1}\right]^2 , \tag{32}$$

$$\beta=\frac{\Delta_1(V_2^2-V_1^2)}{4\Delta_1^2+V_1^2+V_2^2} , \tag{33}$$

$$\gamma=\frac{\Delta_1V_1V_2}{4\Delta_1^2+V_1^2+V_2^2} . \tag{34}$$

Resulting from the above Eqs. (31)–(34), it can be shown that the optimal detunings are determined with the following relations:

$$\frac{T_0V_1V_2}{\sqrt{2}\Delta_1}=\pi , \tag{35}$$

$$\Delta_2=\frac{\Delta_1(V_2^2-V_1^2)}{4\Delta_1^2+V_1^2+V_2^2} . \tag{36}$$

Consequently, Ω_1 and Ω_2 have been defined for an optimal detuning. The homogeneous broadening width related to an excitation process can be obtained from Eqs. (31)–(34). Really, if $\Delta_2=\beta\pm\gamma$ the amplitude of ρ_{33} becomes 0.5, so the homogeneous bandwidth is $V_1V_2/2\Delta_1$.

Since the optimal value of Δ_1 is given by Eq. (35), it is easy to understand that the homogeneous bandwidth is $\pi/\sqrt{2}T_0$ s^{-1}. Therefore, since the bandwidth of the excitation process is very narrow, it is expected that the selectivity of the excitation will be very high.

III. APPLICATIONS

Now we can choose some photoionization schemes for the purpose of concrete examination of the above optimal-detuning technique. First, we take a scheme suitable for a selective photoionization of ^{168}Yb. The scheme is presented as $6\,^1S_0$ (0 cm^{-1}) $\rightarrow 6\,^3P_1$ (17 992 cm^{-1})$\rightarrow(\frac{7}{2},\frac{3}{2})_2$ (35 192 cm^{-1}) [4]. The dipole moments of the transitions are known to be $d_1=2.7\times10^{-30}$ C m and $d_2=4.3\times10^{-31}$ C m. The laser pulses are supposed to be of square shape with the half duration time T_0 equal to 10^{-8}s and with energy density of 0.5 mJ/cm^2. In this case, the Rabi frequencies for the first and the second transitions V_1 and V_2 are 1.76×10^9 and 2.81×10^8 rad/s, respectively. Using the formulas of Eqs. (35) and (36) we get the optimal excitations when $\Omega_1=4741$ MHz and $\Omega_2=-4888$ MHz. The population dynamics is shown in Fig. 2, where the analytic result of Eq. (25) is denoted by a thick solid curve, and the numerical one of Eqs. (2)–(14) is represented by three thin solid curves. Comparing the results of the third-level population in the figure, it can be seen that they are nearly the same although the numerical curve is slightly shifted form the center. Their maximum values are 99.98% and 97.10%, respectively. Therefore it is revealed that the modified system expressed by Eqs. (16) –(24) is appropriate for calculations of the three-level medium near a two-photon resonance region. In particular, since when detunings are large it is very difficult to calculate Eqs. (2)–(14) numerically, the set of equations is very useful for easy and quick calculations.

For the examination of atomic Ca, we consider the well-known ladder configuration $4\,^1S_0\rightarrow4\,^3P_1\rightarrow5\,^3S_1$.

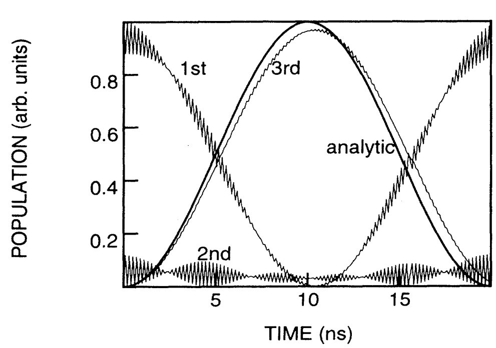

FIG. 2. Atomic-level populations for a selective photoionization scheme of Yb. The three thin solid curves represent the numerical result, while the thick solid curve is the third-level population obtained from the analytic expression. Square pulses with the half duration time of 10 ns and the energy density of 0.5 mJ/cm^2 were used.

The dipole moments of the first and the second transitions are known to be 0.15×10^{-30} and 26.5×10^{-30} C m, respectively. So the rate d_2/d_1 is approximately equal to 187. The optimal detunings are found to be that $\Omega_1=18.89$ GHz and $\Omega_2=14.98$ GHz, and the maximum population of the third level from Eq. (25) becomes 86%. Hence it turns out that the detuning control is a very efficient method for the optimization of the photoionization process of a three-level medium.

Now consider an application of the method to a Doppler-broadened medium. Particularly in the case of collinear waves the situation is more complicated because the detunings are expressed by

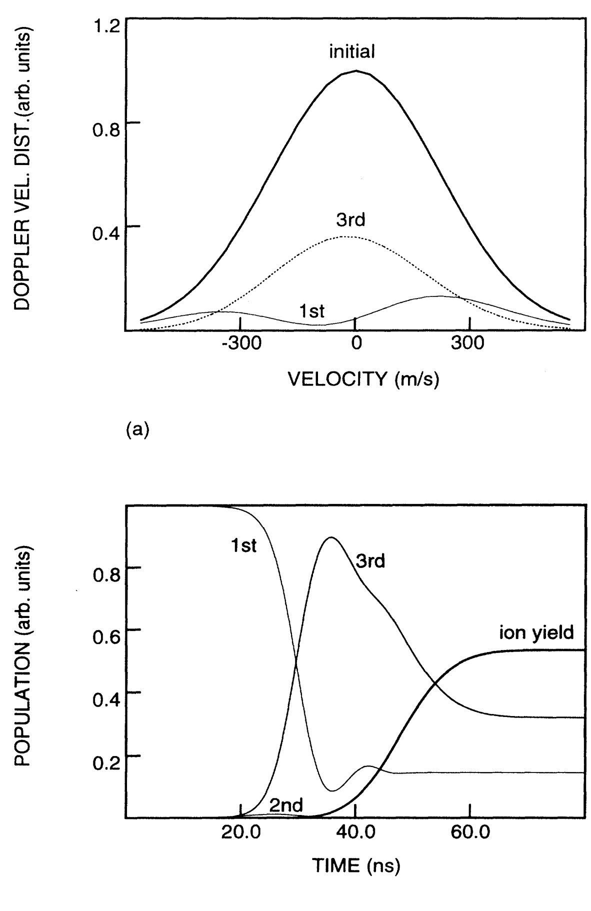

FIG. 3. Doppler velocity distributions and atomic-level populations. Gaussian-shaped pulses with a characteristic time of 10 ns and with the energy density of 0.5 mJ/cm^2 were used. The first and the second lasers were supposed to be counterpropagating, and the time delay of the third laser was 20 ns. The Doppler velocity v_0 was 312 m/s. $\Omega_1=6760$ MHz and $\Omega_2=-6863$ MHz. The Doppler velocity distributions and the atomic-level populations are presented in (a) and (b), respectively. In (a), the thick solid curve is the initial distribution, and the thin solid curve and the dashed one denote the results of the first- and the third-level distributions after the laser-atom interaction.

$$\Omega_1=\omega_{21}\left[1-\frac{v_y}{c}\right]-\omega_1 ,$$

$$\Omega_2=\omega_{32}\left[1-\frac{v_y}{c}\right]-\omega_2 .$$

In this case if we take optimal detunings for a moving atom with v_y equal to 0, the detunings for other atoms with nonzero Doppler velocity cannot be optimized as

$$\Omega_1=\Omega_1^{\rm opt}-\omega_{21}\frac{v_y}{c} ,$$

$$\Omega_2=\Omega_2^{\rm opt}-\omega_{32}\frac{v_y}{c} .$$

Hence it is known that the optimal-detuning condition cannot always be found for all atoms in a Doppler-broadened medium. In the case of collinear waves, the Doppler distribution function $D(v_y)$ will include a very narrow gap such as

$$\delta v_y=\frac{\lambda_1\lambda_2}{T_0(\lambda_1+\lambda_2)\sqrt{2}} ,$$

where λ_1 and λ_2 are the wavelengths of the first and the second transition, respectively. For a transition by a laser pulse in the visible spectrum of light and with a characteristic time of 10^{-8} s, the width of the gap will be approximately 30m/s. So the integral efficiency for all atoms will not be high.

However, in the case of counterpropagating waves, we can expect a better result because the Doppler shifts on the first and th second transition are partly compensated. The detuning from a two-photon resonance is

$$\Delta_2(v_y)=\Delta_2^{\rm opt}+\frac{1}{\sqrt{2}}(\omega_{21}-\omega_{32})\frac{v_y}{c} .$$

If the first and the second transition energies, i.e., ω_{21} and ω_{32}, are close to each other such that $(\omega_{21}-\omega_{32})v_y/c \ll \pi/T_0$, then $\Delta_2(v_y)$ can be approximately $\Delta_2^{\rm opt}$ for all the atoms in a Doppler-broadened medium. We should notice that this method is well known in the atomic-spectroscopy technique, where a Doppler-free line is permitted in transitions.

Results of a numerical study for the inversion of a three-level Doppler-broadened Yb medium are shown in Figs. 3(a) and 3(b). The Doppler velocity $v_0=312$ m/s was used. We approximated the pulse of a real laser system by a Gaussian-shaped one such as

$$I(t)=\frac{U_0}{\sqrt{\pi}T_0}\exp[-(t/T_0)^2]$$

with the characteristic time T_0 equal to 10 ns and the energy density U_0 being 0.5 mJ/cm^2, the same for the three laser pulses. Optimal detunings such as $\Omega_1=6760$ MHz and $\Omega_2=-6863$ MHz were chosen specially for these pulse characteristics. The photoionization pulse was delayed by 20 ns. In Fig. 3(a) the Doppler velocity distributions before and after the laser pulse propagation are shown. In Fig. 3(b) are shown the dynamics of level populations and ion concentration. The photoionization cross section σ_3 was taken to be 6.7×10^{-16} cm^2 [4]. Hence it is known that the optimal-detuning method with counterpropagating waves will give us a relatively high efficiency compared with the result for the case of collinear propagation.

IV. CONCLUSION

A principle for the effective excitation of a three-level medium in selective photoionization has been examined, and a formula for optimal detunings where almost all the atomic population can be inverted into the third bound level has been suggested in the case of a stationary field. The formula depends on the parameters of the laser-atom system. It is found that optimization of the excitation process of a three-level medium with this formula will enable us to enhance the efficiency of photoionization of wanted atoms in the medium as well as to obtain high selectivity.

For the known photoionization schemes of Yb and Ca, the maxima of third-level populations at the optimal detunings have been obtained to be about 100% and 86%, respectively Even in the case of a Doppler-broadened atomic medium, it turns out that the optimal-detuning process is suitable with counterpropagating waves. In particular, some scheme of Yb has the possibility to yield high efficiency in selective ionization of an isotope.

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