Two photon ionization of condensate atoms

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The efficient photoionization of a Bose-Einstein condensate requires the creation of ions with the smallest possible transfer of atoms from the condensate into the thermal phase. The spontaneous decay from excited states into atomic states with momentum different from the initial one should be reduced. We investigate theoretically the two-photon ionization of a rubidium condensate using near resonant excitation to the 6P state and second photon at 421 nm or 1002 nm into the continuum. Different ionization schemes with coherent control of the first excitation and reduction of the spontaneous decay are presented.

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I. INTRODUCTION

The realization of Bose-Einstein condensates (BEC) of alkali atom vapors has attracted much interest into new aspects of photon-matter interaction arising from the quantum nature of the atomic sample. Recently attention has been paid to the photoionization of a BEC by monochromatic laser light [1, 2, 3, 4, 5]. The products of the photoionization process (electrons and ions) obey to Fermi-Dirac statistics. Due to the quantum nature of the initial atomic target and the narrow spectral width of the laser ionization source, the occupation number of the electron/ion final states may become close to unity, especially for a laser excitation close to threshold. If this regime is reached the ionization rate could be slowed down by the Pauli blockade, with the occupation of ionized states determined by the balance between the laser ionization of the condensate and the rate of escape of the ionization products from the condensed system. Moreover, the electrostatic interaction between the ions and neutral atoms in the condensate is expected to originate localized deformations in the condensate density distribution. Either an increase or a decrease in the neutral atom spatial density, depending on the sign of the atom-ion phase shift, could be originated in small regions surrounding the ions [6].

The experimental study of these processes requires the production of a large ionic concentration within a condensate on time shorter than the timescale on which the ions escape from the system, of the order of hundred ns for typical condensate samples, without simultaneously destroying the condensate. Instead, in the two-photon ionization experiments reported in refs. [2, 3, 4, 5] the photoionization laser produced a significant transfer of atoms from the condensate into the thermal cloud. Those experimental investigations demonstrated that the rubidium two-photon ionization was not performed in efficient way, i.e. without producing a large depletion of the condensate atoms with their transformation into thermal atoms. Indeed, in typical two-photon ionization experiments the ionization is increased only at the expenses of a large production of thermal atoms.

The present work reports a theoretical investigation of the efficiency realized in the ionization of atomic rubidium. This work addresses a target which is different from those associated with the typical investigations of multiphoton processes. Our aim is not to increase the multiphoton ionization probability per se. In contrast, the production of ions is compared to optical pumping rate, produced by the spontaneous emission, transferring condensate atoms into the thermal cloud. The ionization efficiency will be determined by the decrease in the loss towards atomic states initially not occupied, i.e. a decrease of optical pumping into sink thermal states, and by the increase in the number of ions.

Our condensate ionization scheme takes place in a three-level cascade scheme, the final one being the atomic continuum. The condensate losses are produced by spontaneous emission from the intermediate excited state. An efficient ionization is realized for an atomic transfer into the continuum with a reduced real excitation of the intermediate level, whence a reduced role of spontaneous emission processes from that intermediate level. The task of reducing the transfer towards sink levels is usually reached applying the STIRAP technique for the coherent transfer between the initial and final levels of a three level scheme, as analyzed in [6, 7]. For transfers to and from a continuum as in [8, 9, 10], an efficient STIRAP transfer, which relies on the coherent coupling of the initial and the final state, can be realized only using very short laser pulses, i.e. shorter than the characteristic decoherence time of the system.

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In the case of photoionization, the use of laser pulses shorter than the typical electron-ion decoherence times would prevent from creating the narrow kinetic energy distribution for the electrons and ions required for the Pauli blockade. Instead we will analyze here the two-photon ionization produced by narrow band cw radiation.

For long interaction times the photoionization and optical pumping processes can be described through a simple rate equation approach. At shorter interaction times, comparable to or smaller than the spontaneous emission lifetime from the first excited state, the determination of the photoionization efficiency requires the solution of density matrix equations. Our solution of the density matrix equations searching for a regime of efficient ionization has strong connections with the investigations of refs. [12, 13] for the lineshapes of atomic excitation and ionization in a cascade three-level system under two-color excitation as a function of the time delay in the application of the two lasers.

Section II describes the characteristics of our three-level scheme. Section III analyzes the case of simultaneous application of the excitation and photoionization lasers for long interaction times on the basis of the rate equation solution. In Section IV we analyze the ionization efficiency for different temporal shapes of the excitation and ionization laser pulses using the density matrix approach. The lengths of the laser pulses and their time delay represent the experimental parameters controlling the ionization efficiency. Section V analyzes ionization based on a rapid adiabatic transfer.

II. ATOMIC LEVEL AND HAMILTONIAN

We investigate the rubidium two-photon ionization scheme from the $5S_{1/2}$ ground state to the continuum with intermediate resonant state the $6P_{3/2}$ state at 2.94 eV above the $5S_{1/2}$ ground state as in the experiment of ref. [4]. The two-photon ionization was produced either by two photons a 421 nm or by a two color process, one photon at 421 nm and a second one at 1002 nm. The photoionization cross sections for absorption of one 421 nm or one 1002 nm photon from the $6P$ state have a small difference, not playing an important role for the present investigation of the photoionization efficiency. Instead a larger intensity was available at the 1002 nm wavelength, and the contribution to ionization of the two color process was much larger than the contribution of the one-color photoionization by two 421 nm photons.

The level scheme of Fig. 1 schematizes the two-photon ionization process from the ground state $|g\rangle$ of the condensate to the condensate ionic state $|i\rangle$ with a near resonant excitation of the intermediate level $|e\rangle$. The population of the intermediate state $|e\rangle$ may decay back into the ground state through emission of spontaneous emission photons. For a sample composed by a Bose-Einstein condensate with atoms having a momentum $p = 0$, the spontaneous emission produces ground state atoms with an atomic momentum $p$ different from zero. These atoms in the $p \neq 0$ ground state constitute a thermal component separate from the condensate, to be described through a sink state $|s\rangle$ whose atomic properties are equivalent to those of the $|g\rangle$ state. The $|s\rangle$ state is filled by the spontaneous emission with rate $\Gamma$ from the $|e\rangle$ state. The interaction of the $|s\rangle$ state with the photoionization radiation is the same one of the $|g\rangle$ state. However the photoionization from the $|s\rangle$ state produces ions having an initial momentum $p \neq 0$ and weakly interacting with the condensate. Therefore that photoionization process is not relevant for our target of electrons/ions with a small kinetic energy[3] and it will not be considered into our analysis.

A laser with angular frequency $\omega_1$, Rabi frequency $\Omega_1$ and detuning $\delta = \omega_1 - \omega_{eg}$ acts on the $|g\rangle \rightarrow |e\rangle$ transition, while a laser with angular frequency $\omega_2$ and intensity $I_2$ acts on the $|e\rangle \rightarrow |i\rangle$ transition. The intensity of the first

![FIG. 1: Three-level scheme for the excitation from the condensate ground state to the condensate ionic state through an intermediate $|e\rangle$ state. The thermal sink state $|s\rangle$ is occupied by the spontaneous emission decay from the $|e\rangle$ state.](image-url)
The evolution of the atomic system interacting with the laser radiation is given by the Optical Bloch Equations for the atomic density matrix \[ \rho \] on the Hilbert space spanned by the four atomic states introduced above.

\[
\begin{align*}
\dot{\rho}_{gg} &= \Omega_1 \text{Im} \tilde{\rho}_{eg}, \\
\dot{\rho}_{ee} &= -\Gamma \rho_{ee} - \Omega_1 \text{Im} \tilde{\rho}_{eg} - (\sigma_1 I_1 + \sigma_2 I_2) \rho_{ee}, \\
\dot{\rho}_{ss} &= \Gamma \rho_{ee}, \\
\dot{\rho}_{eg} &= -\frac{\Gamma}{2} \rho_{eg} - i \delta \rho_{eg} - \frac{\Omega_1}{2} (\rho_{ee} - \rho_{gg}), \\
\dot{\rho}_{ii} &= (\sigma_1 I_1 + \sigma_2 I_2) \rho_{ee},
\end{align*}
\]

with the additional condition \( \rho_{eg} + \rho_{ee} + \rho_{ii} + \rho_{ss} = 1 \), where \( \tilde{\rho}_{eg} \) denotes the interaction representation of \( \rho_{eg} \). We have not included into the density matrix equations the excitation and ionization of the sink state \( |s\rangle \) by the two lasers, because those processes do not modify the number of atoms lost or ionized from the condensate phase. The numerical results reported in the following are valid not only for the simple three-level scheme of Fig. 1, but also for the real rubidium level scheme, with the excited state occupation spontaneously decaying to other intermediate states before reaching the final sink state \( |s\rangle \). Both the ions and the losses into the sink states deriving from the condensate component are equivalent in the two cases.

The efficiency of the ionization will be measured through the ratio \( r \) between the atomic population in the sink state and the atomic population in the ionic state, after the end of the laser interaction

\[
r = \frac{\int_0^\tau \rho_{ss}(t) dt}{\int_0^\tau \rho_{ii}(t) dt},
\]

where the time \( \tau \) is longer than the laser pulse length \( \tau \) in order to include also the spontaneous emission losses towards the sink state at the end of the laser-atom interaction. Notice that in this analysis a large (small) value of \( r \) corresponds to a low (high) ionization efficiency.

For any temporal shape of the applied laser pulses, a key parameter for the photoionization efficiency is the laser detuning \( \delta \) from the \( |g\rangle \rightarrow |e\rangle \) transition. For decreasing detunings the two-photoionization process becomes more efficient owing to the large excitation of the intermediate \( |e\rangle \) state; at the same time, however, also the spontaneous emission becomes more efficient. As a consequence, for decreasing detunings the loss rate towards the \( |s\rangle \) state usually increases faster than the ionization rate, resulting in the decrease of efficiency for the condensate atoms photoionization.
III. LONG LASER PULSES

During the simultaneous application of the two lasers, the atomic population decays from the excited state $|e\rangle$ and is lost from the system with total rate equal to $\Gamma_L = \Gamma + \sigma_1 I_1 + \sigma_2 I_2$. Therefore, at every time $t$ the total population in the $|g\rangle$ and in the $|e\rangle$ states decays with a time dependent loss rate equal to $\Gamma_L \rho_{ee}(t)$. In the case in which $\Gamma, \sigma_1 I_1 \ll \Omega_1$ the ionized fraction of population and the fraction lost by spontaneous emission can be estimated by treating the losses as a perturbation of the steady state solution for the Eqs. (1-5) restricted to the $|\langle g|, |e\rangle\rangle$ two-level system. This perturbative solution is

$$\rho_{ee}(t) = \frac{\Omega_1^2}{2\Omega_1^2 + \Gamma_L^2 + 4\delta^2} \exp\left[-\frac{\Omega_1^2}{2\Omega_1^2 + \Gamma_L^2 + 4\delta^2} (\Gamma + \sigma_1 I_1 + \sigma_2 I_2) t\right]$$

(7)

Supposing the two lasers applied for a time $\tau$ the ionic and sink state occupations may be written as

$$\rho_{ii}(\tau) = (\sigma_1 I_1 + \sigma_2 I_2) \int_0^\tau \rho_{ee}^{\text{pert}}(t) dt = \frac{\sigma_1 I_1 + \sigma_2 I_2}{\Gamma_L} \left(1 - e^{-\frac{\tau}{\tau_L}}\right),$$

(8)

$$\rho_{ee}(\tau) = \frac{\Gamma}{\Gamma_L} \left(1 - e^{-\frac{\tau}{\tau_L}}\right),$$

where we have introduced the timescale

$$\tau_L = \frac{2\Omega_1^2 + \Gamma_L^2 + 4\delta^2}{\Gamma + \sigma_1 I_1 + \sigma_2 I_2}.$$ 

(9)

Therefore all the population is lost from the three-level system if the duration $\tau$ of the pulses is much longer than $\tau_L$. In this limiting case, with no atom remaining in the $|g\rangle$ state, the efficiency of the process is given by

$$r_\infty = \lim_{\tau \to \infty} \frac{\rho_{ee}(\tau)}{\rho_{ii}(\tau)} = f.$$ 

(10)

with the $f$ parameter defined by

$$f = \frac{\Gamma}{\sigma_1 I_1 + \sigma_2 I_2}.$$ 

(11)

For instance, at laser intensity $I_1 = 0.66$ W cm$^{-2}$ corresponding to a Rabi frequency $\Omega_1/(2\pi) = 5$ MHz and laser intensity $I_2 = 2000$ W cm$^{-2}$, the $f$ parameter is equal to 53.8.

IV. PULSED EXCITATION

As demonstrated in refs. [12, 13], under the interaction with coherent pulsed radiation the atomic excitation presents different properties than in measurements performed under the cw excitation regime. In the regime of pulsed excitation, the time evolution of populations and coherences becomes very important. For calculating the ionization efficiency it is necessary to perform the numerical solution of the density matrix equations (1-5). In fact, it should be taken into account that the actual occupation $\rho_{ee}$ of the excited state is characterized by Rabi oscillations with effective Rabi frequency $\Omega_{1\text{eff}} = \sqrt{\Omega_1^2 + \delta^2}$. Because the ionization depends on the occupation of that state, the atomic ionized fraction increases in time with oscillations at the same frequency.

Fig. 2 compares the exact solution of the density matrix equations and the perturbation solution given by Eqs. (8). The average ionized and the lost fraction are well described by the perturbative solution. However, due to the oscillatory evolution of the excited state population, the numerical solution of Eqs. (1-5) is required in order to obtain the correct value of the ionization efficiency parameter $r$.

Whenever $\tau \leq \tau_L$, while no more ionization occurs after the duration $\tau$ of the pulse and the total ionized fraction remains equal to $\rho_{ii}(\tau)$, a finite fraction of population $\rho_{ee}(\tau)$ remains in the excited state at the end of the atom-laser interaction and decays by spontaneous emission into the sink state. Therefore for a pulse of duration $\tau$ the ionization efficiency ratio of Eq. (8) is given by

$$r(\tau) = \frac{\int_0^\tau \Gamma \rho_{ee}(t) dt + \rho_{ee}(\tau)}{\int_0^\tau (\sigma_1 I_1 + \sigma_2 I_2) \rho_{ee}(t) dt} = f + \frac{\rho_{ee}(\tau)}{\rho_{ii}(\tau)}.$$ 

(12)
In general $r > f$ and the maximum efficiency of the ionization process corresponds to the minimum value $r_{\infty} = f$.

The oscillating behavior of the ionization process appears also measuring the ionization as function of the detuning $\delta$. Such a behavior is shown by the results of Fig. 3 where the ionized fraction $\rho_{\text{ii}}$ and the efficiency ratio $r$ are plotted as a function of the laser detuning $\delta$ from the $|e\rangle$ excited state, at fixed interaction time $\tau$. Owing to the choice $\tau = 2\pi/\Omega_1$, $\rho_{\text{ii}}(\tau) = 0$, so that $r$ has a minimum (and the efficiency has a maximum) at zero detuning. At larger values of the detunings $\rho_{\text{ii}}$ decreases with oscillations, and $\rho_{\text{ii}}(\tau)$ assumes values dependent on the effective frequency of the Rabi oscillations. The role of the Rabi oscillations is more clear on the plot of Fig. 4(b) for the efficiency ratio $r$.

For fixed pulse duration the oscillating time dependence of the excited state population with detuning-dependent effective Rabi frequency $\Omega_1^{\text{eff}}$ produces an oscillating dependence of the ionized and of the lost fractions, and thus also of the ionization efficiency, as a function of the detuning $\delta/2\pi$. In particular, the large oscillation of the ionization efficiency is determined by the oscillating amount of population remaining in the excited state $|e\rangle$ at the end of the laser pulse and decaying totally by spontaneous emission into the sink state $|s\rangle$. As a consequence, $r$ assumes oscillating values which are always larger than the $f$ value of Eq. (11), and can be equal to that value only for isolated detunings where the effective Rabi frequency $\Omega_1^{\text{eff}}$ produces a $2\pi$ pulse for the applied duration of the atom-laser interaction.

The occupation of the excited state at the end of the laser pulse can be annulled in the regime defined as coherent population return [12, 13]. For the coherent excitation of two-level atomic systems subjected to light pulses of duration $\tau$ and detuning $\delta$ from resonance, the dynamics of the atomic population is adiabatic if the detuning is significantly larger than the Fourier width of the pulse, i.e., if $\delta \gg 1/\tau$. In this case the dressed state occupied at the end of the excitation is the same one as before the excitation. For a smooth pulse at constant detuning, that state coincides with the atomic ground state, so that even if the excited state is populated during the application of the pulse, all the population is adiabatically brought back to the ground state at the end of the process. Therefore no population fraction remains in the excited state, $\rho_{\text{ii}}(\tau) = 0$. This regime of coherent population return does not produces any loss into the sink state at the laser pulse end. Results showing the advantages of this regime in the two-photon ionization are reported in Fig. 4.

We considered several durations of pulses all of them with the shape shown in Fig. 4(a). For each duration of the IR square pulse, the blue intensity had a Gaussian profile such that the intensity decreased to $1/e^3$ of the peak value in correspondence of the IR pulse start and end. In Fig. 4(b) and (c) the ionized fraction $\rho_{\text{ii}}$ and the parameter $r$ of the the ratio between the atomic fraction in the sink state and the ionized fraction are shown, respectively, as a function of the laser detuning $\delta$, for several durations of the laser pulse and at a given blue Rabi frequency. In all cases, the effects of adiabatic excitation are visible, because for each pulse duration there is a minimum detuning necessary to obtain the optimal ratio $r$. The value of the minimum detuning is only slightly sensitive to the Rabi frequency of the transition, because, as pointed out in [12], no power broadening occurs in a scheme of coherent population return. The limiting value of $r$ reached at large detunings corresponds to the minimum value $f$ calculated at the laser intensities.
FIG. 4: Ionization with coherent population return. In a) temporal shapes of the blue/IR laser for a 125 ns time length of the IR pulse. In b) ionic occupation $\rho_{ii}$ and in c) parameter $r$ describing the efficiency ratio versus laser detuning $\delta/2\pi$, in MHz. $\Omega_1/2\pi = 5.0$ MHz, $I_2 = 1500$ W/cm$^2$, $f = 71.7$. Results for different time lengths of the IR pulse, 30, 60, 125 and 250 ns respectively, with the blue pulse length scaled as shown in a) and discussed in the text.

of the numerical analysis.

V. RAPID ADIABATIC PASSAGE

Another excitation scheme allowing an efficient and robust transfer of the population from the ground to the excited state is the rapid adiabatic passage (RAP) technique\cite{17,18}. This technique takes advantage of the population adiabatic evolution in order to transfer almost the whole population from the ground state to the excited state, blocking completely the population return. RAP is produced by a time dependence of the laser detuning, and for an efficient transfer the laser detuning changes from a very large and negative value to a very large and positive value, or vice versa, during the application of the light. The laser frequency experiences a chirp, and the chirp duration should be shorter than the excited state lifetime. Under these circumstances, the lowest energy adiabatic state of the system coincides with the ground state before the application of the light and with the excited state at the end of the process. When this scheme is applied to the lower atomic transition of the rubidium two-photon ionization, it allows to transfer the ground state population to the $|6P\rangle$ state, from where a pulse of IR light, with the time sequence shown in Fig. 5 (left-hand side), produces the ionization shown in Fig. 5 (right-hand side). Because the IR pulse ionizes the excited state atoms and also modifies the energy of the excited state, for any temporal shape of the two laser pulses the largest two-photon ionization is obtained for a particular delay of the IR pulse with respect to the blue laser pulse. For the pulse shapes of Fig. 5 the maximum ionization was obtained when the center of the IR pulse was delayed by $0.02 \mu$s with respect to the center of the blue pulse. A linear chirp of the blue detuning was examined, but the chirp exact shape had a minor influence on the ionization efficiency, provided that the detuning difference between initial and final value was larger than $\sim 100$ MHz. Suppressing the oscillatory behavior of the excited population, the RAP led to an excited state occupation and an ionized fraction larger than those obtained by pulses of the same shape but at constant blue detuning. For the blue laser scanning of Fig. 5 the ionization fraction was larger by a factor 2 than produced at constant blue detuning (0.0057 against 0.0027). The RAP technique is thus suitable for producing the maximum possible fraction of ionized population within a laser pulse sequence of short duration, during which the ratio between the atomic population in the sink state and the atomic population in the ionic state corresponds to
FIG. 5: Ionization using RAP from the ground state to the excited state. In left plot the temporal shapes of the blue (dotted line) and IR (dashed line) laser pulses and of the blue laser detuning (continuous line) $\delta/2\pi$, in MHz, at $\Omega_1/2\pi = 30.7$ MHz and $I_2 = 2000$ W/cm$^2$. In right plot time evolution of the excited state occupation (solid line) and of ionized fraction excited state (dashed line).

the optimal value $f$. Due to the suppression of the coherent population return, on the other hand, a large fraction of population typically remains in the excited state at the end of the laser pulse and decays into the sink state thereafter. As a consequence, in RAP at long interaction times the efficiency parameter $r$, calculated according to Eq. 6, reaches values larger than those associated to the adiabatic regime. For the parameters considered above the efficiency ratio $r$ was 165 against the minimum value $f = 53.8$.

VI. CONCLUSION

We have examined a problem to be classified as a quantum control one: reach a specific target, the ionization of the condensate, minimizing the losses into a different output channel, the production of thermal atoms. We have explored different two-photon ionization schemes, producing different quantum controls for the atomic excited state. However the quantum control reached for the two-photon ionization is very poor. In effect the ionization process transferring atoms from the excited state to the ionization state is governed by rate equations, whence a classical response instead of a quantum one. Our final result is that the maximum ionization efficiency to be reached through the quantum control on the first excitation, depends on the probabilities of two processes, the spontaneous decay rate and the ionization rate. These rate are not modified within the quantum control of our analysis.

While the qualitative behavior of the ionized fraction depends only on the properties of the laser pulse exciting the first step of the ionization sequence, the absolute value of the ionized fraction depends only on the intensity of the IR light coupling the excited state to the continuum. An analogous statement applies also to the efficiency ratio between the thermal fraction and the ionized fraction. The criteria we have derived for the optimization of the two-photon ionization have a general validity, whereas the absolute numbers we have discussed correspond to the intensities available in the analyzed experiment. The fraction of ionized atoms in these conditions is much smaller than the number of photons scattered by each atom. The application of the optimization criteria we have derived, however, together with IR intensities larger than those we have considered, will allow to reach experimental conditions leading to an efficient large photoionization of Bose-Einstein condensates.

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