Abstract. We calculate the ionization lifetime of rubidium atoms trapped in an intense light field as a function of wavelength and principal quantum number $n$. The ionization lifetimes of ground state atoms range from essentially infinite to less than one second, for example in a 532 nm laser field with intensity $10^6 \text{ W cm}^{-2}$. The ionization lifetime of Rydberg states can be extremely short, e.g. the lifetime of the 35d state in a CO$_2$ laser field with intensity $10^5 \text{ W cm}^{-2}$ and wavelength $10.6 \mu \text{m}$ is of order 100 ns. These results have important implications for achieving long coherence times, and for entanglement schemes based on Rydberg excitation in far-detuned optical lattices. In particular, fast photo-ionization could seriously compromise the possible realization of quantum gates based on Rydberg excitation of alkali atoms in a CO$_2$ laser lattice.

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1. Introduction

The lifetime and coherence time of atoms in optical dipole traps and optical lattices is important in a wide range of research areas including Bose–Einstein condensation (BEC) [1]–[6], degenerate Fermi gases [7]–[10], optical clocks [11], and quantum information processing (QIP) [12]–[16]. The main limitations to the lifetime are heating due to spontaneous emission which can be reduced by detuning far from resonance [17, 18], and collisions with the background gas which are reduced by using ultra-high vacuum (see for example figure 37 in [17]). For the extreme case of far detuning and low background gas pressure, coherence times of a few seconds [19] and lifetimes of a few hundred seconds have been demonstrated [7]. In this limit where high intensity is required to provide sufficient trap depths, other processes such as photo-ionization could become important, particularly in cases where the atoms are excited to high lying energy levels. The ionization lifetime of highly excited states in optical lattices is important for experiments aiming to realize quantum gates based on the dipole–dipole interaction between Rydberg atoms [20]–[22]. In addition, photo-ionization of ultra-cold atoms [23] is of intrinsic interest in the context of charge hopping [24] and the interactions between ions and quantum degenerate gases [25, 26].

In this paper, we calculate the ionization lifetime of alkali atoms in a far-off-resonance optical trap as a function of the trap wavelength and principal quantum number $n$ of the atomic state. The calculations are performed using a model potential [27]. This approach has been previously used to model two-photon ionization [23], to calculate photo-ionization cross-sections [28], and to calculate dynamic polarizabilities [29]. Because of the large quantum defect, the low-$l$ states of the heavy alkalis often have an ionization lifetime differing significantly from that of the corresponding states of atomic hydrogen. Their photo-ionization cross-section can be estimated semi-analytically within the quantum-defect theory; however, an entirely numerical calculation is more convenient [30]. For the model potential adopted here, the computation is easily performed with existing library programs based on the Floquet formalism [31]. We have therefore used this method. At the relatively weak intensities involved in optical lattices, the calculation is equivalent to a perturbative calculation of the photo-ionization rate from Fermi’s Golden Rule.

Here, we focus on rubidium atoms as they are most commonly used in experiment on BEC and QIP. However, we expect that all the main conclusions of the paper also apply to the other alkalis. In particular, we find that the ionization lifetime of highly excited d-states of hydrogen, lithium, sodium, potassium and caesium atoms in a CO$_2$ laser field is as short as or even shorter than that of rubidium atoms.

While a great deal of work has been devoted to the photo-ionization of alkali atoms in their ground state or in their low excited states, photo-ionization of Rydberg states seems to have received little attention so far. We are aware of only three previous investigations of the photo-ionization of Rydberg states of rubidium, namely the calculations of Aymar et al [28] and of Saffman and Walker [22], and the recent measurement by Gabbanini of the photo-ionization cross-sections of nd states ($n = 16$–20) at the CO$_2$ laser wavelength [32]. Aymar et al [28] calculated the photo-ionization at threshold for ns, np and nd states with $n$ up to 20, using a similar central-field model as adopted here. Saffman and Walker give the cross-section for the photo-ionization of high Rydberg states at 1.01 $\mu$m wavelength, which they obtained using a different model potential. Where comparison is possible, our results are in agreement with these previous theoretical results and recent experimental data$^2$ [32].

$^2$ For d states our cross-sections at threshold reproduce those obtained by Aymar et al [28] for their potential (ii).
Table 1. Binding energy of some of the bound states supported by the model potential and centroids of the experimental fine structure multiplets [34], for rubidium, in cm$^{-1}$. The experimental value quoted for the 27s state is extrapolated from lower levels using the quantum defects quoted in [35].

| State | Model | Exp. | State | Model | Exp. |
|-------|-------|------|-------|-------|------|
| 5s    | 33697 | 33691| 6d    | 5021  | 5003 |
| 5p    | 20909 | 20993| 6s    | 13573 | 13557| 7d    | 3421  | 3410 |
| 4d    | 14313 | 14336| 5d    | 8019  | 7989 |
| 6s    | 9960  | 9937 | 7p    | 5855  | 5839 |
| 6p    | 3421  | 3410 | 7s    | 193   | 193 |
| 7p    | 3196  | 3192 | 27s   | 193   | 193 |

The paper is set out as follows: in section 2, we introduce the model potential method and discuss the calculation of the ionization lifetime. In section 3, we present results of the calculations, and finally in section 4, we summarize the main results.

2. Model potential and calculation of the ionization lifetime

In the simple non-relativistic model adopted here, we treat the core as inert and represent its interaction with the valence electron by a central potential, $W(r)$, of the type introduced by Klapisch [27]. Namely, in atomic units,

$$W(r) = -[1 + A \exp(-\alpha r) + B r \exp(-\beta r)]/r,$$

with the parameters $A$, $B$, $\alpha$ and $\beta$ optimized, so that the energy levels of the potential match the experimental energies of the real atom (the centroids of the spin–orbit multiplets) as closely as possible. For rubidium, we use $A = 36$ au, $B = 6.43527$ au, $\alpha = 3.33355$ au and $\beta = 1.3698$ au [33]. For the other alkalis, we use the parameters originally proposed by Klapisch [27]. The parameters adopted for rubidium ensure a closer agreement of the 5s level with the exact ground state energy. This level and a few other ones are compared to their experimental positions in table 1. They differ from the latter by 10–20 cm$^{-1}$ for most of the lowest lying states, and by less than 0.2 cm$^{-1}$ for the 26p, 25d and 27s states. The largest difference, 84 cm$^{-1}$, is found for the 5p state. Still, this level falls in between the two fine structure components of the real 5p state.

We assume that the laser field is linearly polarized and can be described classically. Taking the $z$-axis along the direction of polarization, the model Hamiltonian reads

$$H(t) = H_0 - e F(t) z,$$

with $H_0$ the field-free Hamiltonian, $F(t)$ the electric field of the laser and $e$ the charge of the electron. We assume that the field is monochromatic and set $F(t) = F_0 \sin(\omega t)$, where $\omega$ is the angular frequency of the laser. Under its effect, the stationary bound states of the field-free atom...
turn into quasistationary, decaying dressed states. They are described by wavefunctions of the Floquet form [36, 37]

\[ \Psi_j(r, t) = \exp(-i\epsilon_j t/\hbar)F_j(r, \omega t), \]  

where the functions \( F_j(r, \omega t) \) are periodic in time with the same period as \( F(t) \). These wavefunctions must satisfy radiation (Siegert) boundary conditions in order to represent an atom initially in a bound state and decaying by photoionization. These conditions restrict the quasienergies \( \epsilon_j \) to discrete complex values. For each dressed state, one can write

\[ \epsilon_j = E_j + \Delta_j - i\Gamma_j/2, \]  

with \( E_j \) the energy of state \( j \) in the absence of field, \( \Delta_j \) the ac-Stark shift of this state, and \( \Gamma_j \) its ionization-induced energy width. Since \( |\Psi_j(r, t)|^2 \propto \exp(-\Gamma_j t/\hbar) \) averaged over one optical cycle, the ionization lifetime of state \( j \) is simply \( \hbar/\Gamma_j \).

The quasienergies of the dressed states can be found by solving the Schrödinger equation for a wavefunction of the form

\[ \Psi(r, t) = \exp(-i\epsilon t/\hbar)\sum N \exp(-iN\omega t)\psi_N(r), \]

where the index \( N \) characterizes the number of photons (real or virtual) absorbed or emitted by the atom. It is useful to expand the dipole operator similarly,

\[ -eF(t)z = V_+ \exp(-i\omega t) + V_- \exp(i\omega t), \]

and to recast the Schrödinger equation into the following system of coupled differential equations,

\[ (\epsilon + N\hbar\omega - H_0)\psi_N(r) = V_+\psi_{N-1}(r) + V_-\psi_{N+1}(r). \]

These equations can be solved nonperturbatively. However, this is impractical for the relatively weak laser fields considered here as the ionization widths of the states of interest are smaller than the numerical inaccuracies of such a calculation. Instead, it is appropriate to solve equations (7) in Rayleigh–Schrödinger perturbation theory [37, 38]. We write \( \epsilon = \epsilon^{(0)} + \epsilon^{(2)} + \cdots \), where \( \epsilon^{(0)} \) is of zeroth order in \( F_0 \) and \( \epsilon^{(2)} \) of second order in \( F_0 \). To zeroth order in the field, we set \( \epsilon^{(0)} = E \), \( \psi_0(r) \equiv \phi(r) \), and \( \psi_N(r) \equiv 0 \) for \( N \neq 0 \), with \( E \) an eigenenergy of the field-free Hamiltonian and \( \phi(r) \) the corresponding eigenfunction. Hence, to first order in the field we have

\[ (E \pm \hbar\omega - H_0)\psi_{\pm 1}(r) = V_\pm \phi(r), \]

from which follows

\[ \epsilon^{(2)} = \int \phi(r)^*V_+\psi_{-1}(r) \, dr + \int \phi(r)^*V_-\psi_1(r) \, dr \]

to second order in the field. This procedure can be iterated to obtain the quasienergy to higher order, which is useful, for instance, for estimating ionization lifetimes at wavelengths where one-photon ionization is impossible. The perturbative photo-ionization rates obtained in this way are identical to those which would be obtained from the Golden Rule and its
multiphoton generalization [39]. We have checked that the perturbative quasienergies converge to the nonperturbative ones at intensities large enough for the latter to be calculable. We solve these equations by expanding the harmonic components on a discrete basis of spherical harmonics \( Y_{lm}(\hat{r}) \) and of complex radial Sturmian functions,

\[
S_{\nu l}(\kappa, r) = N_{\nu l}(2\kappa r)^{|\nu|}e^{-\kappa r}F_1(1 - \nu, 2l + 2, 2\kappa r),
\]

where \( N_{\nu l} \) is a normalization coefficient [31, 37, 39]. The calculation reduces thereby to solving linear algebraic equations for the coefficients of the expansion, which can be tackled by standard numerical methods. The boundary conditions are implemented by giving a small negative imaginary part to the parameter \( \kappa \) of the Sturmian functions [40]. The radial basis is taken large enough that the ionization width remains stable for reasonable variations of \( \kappa \). Because of the disparity of length scales between the short-range part of the model potential and the spatial extension of Rydberg states, in some cases as many as 600 Sturmians were required in each partial wave to achieve convergence.

3. Results

All the results presented here have been calculated for a laser field intensity of \( 10^5 \) W cm\(^{-2}\). This intensity corresponds to a 4 W laser beam focused to a 50 \( \mu \)m waist and is chosen to represent a typical intermediate intensity used in experiments on far-red detuned optical traps. For far-blue detuned trap where the atom is trapped at low intensity significantly lower intensities may be typical. It is straightforward to scale the results for any intensity as explained below.

The excited states of rubidium above 12d can ionize by absorption of a single photon over the complete range of wavelengths considered here (up to 10.6 \( \mu \)m, the wavelength of a typical CO\(_2\) laser field). The atom may absorb more than one photon too, but in these relatively weak fields the rate of this process is smaller by several orders of magnitude. The perturbative calculation can thus be limited to second order in \( F_0 \), and the ionization lifetime of these Rydberg states is inversely proportional to the intensity of the laser field. The lifetime of a few highly excited d states is shown in figure 1 for selected wavelengths between 430 nm, which is close to the 5s–6p resonance at 421 nm and 10 600 nm. (The data points have been joined by straight lines to guide the eye. The lifetime is affected by resonances with other bound states and does not vary smoothly between some of the calculated points.) As seen from the diagrams, the ionization lifetime of these states increases with the principal quantum number and with \( |m| \). As is the case for many species [28, 41], however, it is much shorter in the infrared than in the visible. For example, in a CO\(_2\) field of \( 10^5 \) W cm\(^{-2}\) intensity, the lifetime of the 13d\(_{m=0}\) state of rubidium is of the order of one nanosecond, while that of the 35d\(_{m=2}\) state is of the order of tenth of a microsecond. By contrast, the ionization lifetime of the d states exceeds their natural lifetime below 1000 nm for an intensity of \( 10^5 \) W cm\(^{-2}\). As noted above, these ionization lifetimes are inversely proportional to the intensity. Thus, at an intensity \( 10^6 \) W cm\(^{-2}\), ionization dominates over spontaneous emission for any trapping wavelength above 430 nm. At lower intensity, \( 10^4 \) W cm\(^{-2}\) the lifetime of the nd states (for \( n > 35 \)) increases to greater than a microsecond.

The main result of this paper is the short lifetime of Rydberg atoms in a far infra-red field. This fast photo-ionization compromises the realization of Rydberg gates [20, 21] in for example a quasi-electrostatic lattice based on a CO\(_2\) laser [29]. However, the short lifetime could be an advantage in experiments where fast ionization of low \( n \) Rydberg states (that are difficult to ionize using electric fields) is required as part of a detection scheme [42].

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Figure 1. Ionization lifetime of excited states of Rb in a laser field of $10^5 \text{ W cm}^{-2}$ intensity. (a) Lifetime of the 13, 19, 25 and $35\,\text{d}_{m=0}$ states for two different wavelengths, 430 and 10 600 nm. The crosses indicate the natural lifetime of these states [35]. (b) Lifetime of the $13\,\text{d}_{m=0}$ and $35\,\text{d}_{m=0}$ states (●), of the $13\,\text{d}_{m=2}$ and $35\,\text{d}_{m=2}$ states (○) and of the $37\,\text{s}$ state (■) for five different wavelengths, 430, 532, 1064, 2000 and 10 600 nm.

The recent measurement of the photo-ionization cross-section of highly excited d states of rubidium at 10.6 $\mu\text{m}$ [32] gives an opportunity to ascertain the accuracy of our calculation. The cross-sections for 16d, 17d, 18d, 19d and 20d were measured to be, respectively, 39, 35, 32, 30 and 30 Mb, with an estimated total error around 40%. We obtain, respectively, 30, 26, 21, 18 and 16 Mb for the cross-sections averaged over the magnetic sublevels, and 50, 43, 36, 30 and 26 Mb for ionization from the $m = 0$ sublevel alone. (The cross-sections at threshold are significantly higher [28].) Given the uncertainty in the distribution between the different magnetic sublevels in the experiment, our results are consistent with the data.

Owing to a Cooper minimum in the photo-ionization cross-section, the ionization lifetime of the s states of rubidium does not increase monotonically with frequency [28]. We find that this minimum is located too far from the threshold to affect significantly ionization in the far infra-red: while in the near infra-red and in the visible the ionization lifetime of s Rydberg states exceeds that of neighbouring d state by orders of magnitude, the difference is typically of the order of a factor 10 for a CO$_2$ field (figure 1).

In figure 2, rubidium is compared to other alkalis and to atomic hydrogen for the case of a CO$_2$ laser field. As seen from the diagrams, the short ionization lifetimes in the far infrared noted above are not specific to rubidium. For d states, the calculation predicts even shorter ionization lifetimes for the other alkalis and for hydrogen. As for rubidium, the cross-section for photo-ionization from s states of sodium, potassium and caesium passes through a Cooper
Figure 2. Ionization lifetime of various states of atomic hydrogen (○), lithium (×), sodium (+), potassium (□), rubidium (●) and caesium (△) in a laser field of 10³ W cm⁻² intensity and 10.6 μm wavelength. Solid lines: quasiclassical results for atomic hydrogen. The horizontal scale shows the effective principal quantum number of the states, \( n^* = n - \delta \), where \( n \) is the principal quantum number and \( \delta \) the quantum defect [35]. (a) \( l = 0 \) states, (b) \( l = 2, m = 0 \) states, (c) \( l = 2, m = 2 \) states.

Minimum at shorter wavelengths, which increases their ionization lifetime significantly. Lithium is exceptional in that the cross-section for photo-ionization from s states is a minimum at the threshold [28]. Compared to hydrogen, this minimum increases the ionization lifetime of the low Rydberg states in a CO₂ field by about two orders of magnitude. It is only for this element that we find lifetimes in the far infra-red in excess of 1 μs at 10⁵ W cm⁻² when \( n \leq 40 \).

The photo-ionization cross-section of atomic hydrogen can be obtained accurately using quasiclassical methods [30, 43]. The following formulae, valid for \((l + 1)^3\omega \ll 3\) au, result from equations (13) of [30]: with the photo-ionization rate \( R = \Gamma / \hbar \), the electric field amplitude \( F_0 \) and the angular frequency \( \omega \) expressed in atomic units, and \( x \equiv (\pi/2)(\omega/3)^{1/3} \),

\[
R/F_0^2 \approx 0.2651n^{-3}\omega^{-10/3}(1 + x)^2/3
\]  

(11)

for \((n, l = 0, m = 0)\) states,

\[
R/F_0^2 \approx 0.2651n^{-3}\omega^{-10/3}[(1 - 2x)^2(4/15) + (1 + 3x)^2(9/35)]
\]  

(12)

for \((n, l = 2, m = 0)\) states,

\[
R/F_0^2 \approx 0.2651n^{-3}\omega^{-10/3}[(1 - 2x)^2(1/5) + (1 + 3x)^2(8/35)]
\]  

(13)

for \((n, l = 2, m = 1)\) states, and

\[
R/F_0^2 \approx 0.2651n^{-3}\omega^{-10/3}(1 + 3x)^2/7
\]  

(14)
Figure 3. Ionization lifetime of the 5s state of Rb in a laser field of $10^5$ W cm$^{-2}$ intensity at wavelengths between 360 and 594 nm

for $(n, l = 2, m = 2)$ states. The corresponding ionization lifetimes are represented by solid lines in figure 2. They are in close agreement with the exact results. With the exceptions of the s states of lithium and of the d states of caesium, which are markedly non-hydrogenic [28], the lifetimes of the alkalis increase with the effective principal quantum number following the same $n^3$ power law as for atomic hydrogen. However, in most cases the proportionality factor is not the same and scales differently with the frequency.

The ground state of rubidium can ionize by one-photon absorption only at wavelengths below 297 nm. Two-photon ionization is possible up to 594 nm. Except where enhanced by resonances with intermediate p states, however, this is a relatively slow process. The ionization lifetime of this state is shown in figure 3 for wavelengths between 360 nm (close to the 7p resonance) and 594 nm, again for $10^5$ W cm$^{-2}$ intensity: it decreases below 1 s close to the resonance with the 6p state, around 421 nm. The lifetime is of order 10 s close to ionization threshold (594 nm), and is typically much longer above the resonances. It is worth noting, however, that the ionization lifetime is here inversely proportional to the square of the intensity. For example, while the lifetime of the ground state at 532 nm is about 50 s at $10^5$ W cm$^{-2}$, it is only about 0.5 s at $10^6$ W cm$^{-2}$. The ionization lifetime of the ground state is practically infinite at wavelengths above 594 nm (except in the immediate vicinity of resonances) as ionization by absorption of more than two photons is excessively slow in the conditions considered here: At an intensity of $10^5$ W cm$^{-2}$, the model predicts a lifetime of $2.9 \times 10^6$ s at 800 nm and $1.1 \times 10^{13}$ s at 1064 nm.

We note, finally, that at the intensities relevant here, photoionization is likely to be negligible compared to spontaneous decay for any excited state that does not ionize by one-photon absorption. For example, the ionization lifetime of the $5p_{m=0}$ state of rubidium at $10^5$ W cm$^{-2}$ intensity is 71 s at 532 nm and $1.5 \times 10^8$ s at 1064 nm, which of course exceeds the natural lifetime of this state by many orders of magnitude.

4. Conclusions

In summary, we have presented a calculation of the ionization rate of alkali atoms trapped in light induced potentials. The ionization lifetime of ground state atoms is relatively long, except
for intense laser beams with a wavelength below the two-photon ionization threshold (594 nm in Rb). For example, the lifetime of Rb atoms at 532 nm with an intensity of $10^9$ W cm$^{-2}$ is 0.5 s. For Rydberg states, the ionization lifetime can be extremely short in particular in the limit of far red detuned trapping light. For example, the ionization lifetime of Rb Rydberg atoms with $n = 35$ is shorter than the natural lifetime at trapping wavelength above 1 $\mu$m for an intensity of $10^5$ W cm$^{-2}$, and above 430 nm for an intensity of $10^6$ W cm$^{-2}$. The results indicate that careful selection of the principal quantum number will be required to avoid fast photo-ionization in quantum gate schemes based on Rydberg excitation of alkali atoms in a CO$_2$ laser lattice. However, one could turn the fast photo-ionization into an advantage and use intense far-infra red fields to ionize and detect low $n$ Rydberg states that would be difficult to ionize using static electric fields.

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