Optimal control of multiphoton ionization of Rb$_2$ molecules in a magneto-optical trap

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We present optimal control, using femtosecond laser pulses in a closed-loop experiment, of the ionization process of ultracold rubidium molecules formed in a magneto-optical trap. The resulting spectrum of the optimally shaped pulse exhibits two frequency bands, which drive a resonantly enhanced three-photon ionization. The obtained data reveals the ionization process where only the electronic states corresponding to the 5s5d-asymptote are utilized for the second excitation step.

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I. INTRODUCTION

In the past decade, processes in ultracold atomic ensembles have been extensively studied and many fascinating effects have been discovered. Expanding these investigations towards ultracold molecules in the gas phase is a logical step to gain further knowledge about fundamental quantum dynamics. Transferring the techniques of atom cooling and trapping directly to molecules is not possible due to their internal degrees of freedom [1]. Techniques available to obtain cold molecules include buffer gas cooling [2], Stark decelerators [3], or Feshbach resonances [4]. Another approach is photoassociation of ultracold atoms in magneto-optical traps (MOTs) [5] which already has been successfully experimentally applied using cw lasers with narrow bandwidths [6,7]. It is proposed that a more efficient way of generating deeply bound cold molecules could be achieved with pulsed laser photoassociation [8], and this was particularly investigated for the rubidium system [9].

Lasers with pulse lengths shorter than the oscillation period of molecules are a widespread tool for investigating their internal dynamics. By manipulating the spectral components of femtosecond laser pulses the electrical field can be modulated in time leading to a complex structure tailored to the particular system [10]. Controlling such a laser field by evolutionary algorithms is a standard tool in the field of femtochemistry. By implementing an experimental feedback loop it becomes feasible to control reactions and drive the quantum system to desired states [11]. Many of these so-called coherent control experiments together with the analysis of the obtained pulse form lead to a deeper understanding of the involved processes [12] that are particularly successful on small alkali clusters [13]. Merging the techniques of laser cooling with the methods of coherent control opens a fascinating field in physics with a plethora of possibilities. The first experiments of this type were the suppressing of molecule formation by chirped femtosecond laser pulses [14] and optimal control of the enhancement of excitation of Rb$_2$ [15].

In this paper, we present results of the ionization of rubidium dimers in a MOT by manipulating the spectral components of femtosecond laser pulses in a closed feedback loop optimization. A supplementary experiment, using a notch filter, supports the results of the optimization. The resulting spectra are analyzed in order to understand the underlying processes.

II. EXPERIMENTAL SETUP

In our experiments, $^{85}$Rb atoms are captured in a background vapor loaded magneto-optical trap at temperatures in the order of 100 $\mu$K. The density in normal MOT configuration where the repumper is set to the $F=2 \rightarrow F'=3$ hyperfine transition is about $10^{10}$ atoms/cm$^3$ [16]. The trap can also be operated as a detuned dark spontaneous-force optical trap (dark SPOT) [17] with densities of about $10^{11}$ atoms/cm$^3$ when 90% of the atoms are kept in the $F=2$ ground hyperfine state by detuning the repumping laser 30 MHz to the $F=2 \rightarrow F'=3$ hyperfine transition and by physically blocking the core part of the repumping laser beams. Molecules are continuously formed in the trap by either three body collisions or photoassociation through the trap lasers [18,19]. The femtosecond laser pulses are produced by a Coherent Mira oscillator with a power of 400 mW and 76 MHz repetition rate. These pulses are amplified by a Coherent RegA 9050, delivering pulses of 3.5 $\mu$J energy at a repetition rate of 250 kHz corresponding to an average power of 875 mW at a central wavelength of 780 nm with 23 nm bandwidth. These pulses are shaped by a 2 $\times$ 640 pixel liquid crystal modulator (CRi, SLM-640) which is located in the Fourier plane of a zero dispersion compressor that is able to manipulate the spectral phase and amplitude simultaneously and independently. The zero dispersion compressor is built in 4/f-configuration with gratings of 2000 groves per mm and cylindrical lenses each with 250 mm focal length, which leads to a resolution of 0.14 nm/pixel (2.3 cm$^{-1}$/pixel). Evolution strategies are used to operate the pulse shaper based on the molecular ion signal [20]. The atomic and molecular ions produced in the trap are extracted by a constant electrical field of 40 V/cm
and are detected by a channeltron staged after a quadrupole mass filter.

III. OPTIMIZATION OF THE IONIZATION PROCESS

At first, the ion yield in the dark SPOT is optimized in a closed loop experiment using a parametric algorithm. This algorithm optimizes four Gaussian peaks in the spectral amplitude of the laser pulse whereas the phase pattern is freely optimized [21]. The parameters of each Gaussian peak are the positions that can be shifted within the limits of ±15 nm around the central wavelength, the width, which can be varied from 0 to 0.55 nm (0 to 9 cm⁻¹), and the amplitudes with a transmission range from zero to one. By overlapping peaks or setting the width or transmission to zero the algorithm can reduce the number of spectral peaks. Peaks with a larger width can be produced by setting peaks side by side. The spectrum of the resulting pulse is less complex and easier to interpret than the one obtained by free optimization of the amplitudes and reveals only the most important frequencies. Another advantage of parametric modulation in the frequency domain is the smaller number of parameters compared to free optimization, which restricts the algorithm to a subset of the search space and therefore it converges faster. This shortens the experiment to a feasible duration because the low detection rate requires a long averaging time of 16 s per individual resulting in 8 min per generation.

The Rb₂-ion yield acts as the feedback signal for the algorithm which has to be maximized. It increases (starting from random initial conditions) and converges after about ten generations, as depicted in Fig. 1. The Rb₂⁺ count rate obtained with the optimized pulse is enhanced by a factor of about 17 compared to an unshaped short pulse of the same power (22 mW). The spectrum of the obtained optimal pulse is shown in Fig. 2. It exhibits two frequency bands at 12 815 and 12 883 cm⁻¹ (780.4 and 776.2 nm). This phase and amplitude shaped pulse is strongly modulated in time and about 11 picoseconds long. The four Gaussian peaks are reduced to these two frequency bands by the algorithm. Thus other frequencies seem to be irrelevant or even obstructive for the ionization process. The width of optimized pulses may allow information about the vibrational distribution of the ultracold molecules formed in the MOT to be inferred, if the spectral resolution would be high enough.

The molecular formation rate in the MOT depends on the atomic density [18]. It is reduced by the femtosecond laser pulse due to atomic ionization and light scattering forces. Therefore the formation and ionization of molecules must be balanced with the atomic depletion, which are both induced by the femtosecond laser pulse.

This experiment was repeated using only the oscillator, which has a lower pulse energy (nJ compared to μJ) but a higher repetition rate, so that the mean power is comparable. With these pulses the obtained ion count rate using the optimized spectrum is also enhanced compared to no shaping, but the molecular count rate is seven times smaller than in the high intensity case. This is consistent with a multiphoton ionization, which is a nonlinear effect, where high peak intensities are more efficient.

IV. FREQUENCY FILTER SCANS IN THE FOURIER PLANE

For a further survey of the relevant spectral components responsible for cold molecule ionization, a supplementary experiment was carried out. Instead of selecting frequency bands to include we systematically removed parts of the spectrum.

A needle in the Fourier plane of the shaper acts as an adjustable optical notch filter with 100% attenuation (the shaper results in approximately 2% leakage). This needle has a diameter of 1.5 mm, is moved by a micrometer stage, and blocks a band 36 cm⁻¹ wide. In this experiment we are mainly interested in the interaction of the frequencies around the two important bands from the previous experiment. The frequencies beneath 12 599 cm⁻¹ (including the atomic D₁ line) are thus blocked by a knife edge in the Fourier plane (see inset of Fig. 3) to reduce trap loss. Furthermore we attenuated the femtosecond laser beam to an average power of 88 mW in the interaction region.

For each needle position 1000 samples of the ion count rate and of the fluorescence signal are taken and the average
value is plotted in Fig. 3. These measurements were carried out in dark SPOT and also in normal MOT configuration, producing qualitatively similar results.

The graph shows a constant signal apart from the needle positions at the frequencies around 12 815 cm$^{-1}$ (780.4 nm) and 12 883 cm$^{-1}$ (776.2 nm), where the Rb$_2$-ion signal is lower. The band around 12 883 cm$^{-1}$ seems to have a larger effect on the total molecular ion yield, as the signal plunges practically to zero when it is blocked, compared to the finite rate of approximately 10 Hz, when the 12 815 cm$^{-1}$ is blocked. This applies to both normal and dark SPOT configuration.

The fluorescence shows the opposite behavior. When one of the bands is blocked, it increases by approximately 40%. The only difference between the results obtained with dark SPOT and normal MOT is the fluorescence being four times higher for the latter. The fluorescence is a measure for the steady-state number of excited atoms in the trap. Likely trap-loss mechanisms include resonant excitation of atomic rubidium by the two frequencies followed by ionization, and loss of atoms due to photon pressure. This demonstrates that both frequency bands are needed to decrease the fluorescence of the MOT corresponding to a lower number of trapped atoms.

FIG. 3. (Color online) (a) Ion signal depending on the position of a needle in the Fourier plane. (b) Corresponding fluorescence. The inset shows the associated spectrum at a needle position of 12 903 cm$^{-1}$ (775 nm) and the absence of frequencies beneath 12 599 cm$^{-1}$ (793.7 nm).

V. DISCUSSION

The optimization and the filter scan through the Fourier plane both reveal two frequency bands at 12 815 cm$^{-1}$ (780.4 nm) and 12 883 cm$^{-1}$ (776.2 nm) which are relevant for the ionization. According to this result the ionization of Rb$_2$ from the electronic ground state can be understood as a three step process where only the two frequency bands efficiently contribute. The observed frequency bands can be associated with corresponding transitions in the potential energy scheme which is depicted in Fig. 4. The molecules which are present in the trap are vibrationally highly excited in the ground state, i.e., the population is concentrated within a few wave numbers below the dissociation limit [19].

The frequencies of the band around 12 815 cm$^{-1}$ (780.4 nm) probably excite long-range bound ground state molecules to the 5s5p$^{3+2}$ asymptote (D$_2$ line) with the associated energy potential curves of this electronic state. In the next step, the population is excited by the frequencies of the band around 12 883 cm$^{-1}$ (776.2 nm), which is exactly the energy difference between the 5s5p$^{3+2}$ and the 5s5d-asymptote at 25 700 cm$^{-1}$. The fine structure splitting for the 5s5p$^{3+2}$→5s5d$^{3+2}$ transition, of 0.18 nm (2.96 cm$^{-1}$), is also too small to be clearly resolved in our experiment.

The interesting feature in the needle scan experiment, where the signal goes to zero when the frequency band around 12 883 cm$^{-1}$ is blocked but still some amount is obtained when the band around 12 815 cm$^{-1}$ is blocked, may be understood within our ionization scheme as being a side-
effect of the molecular production mechanism within a MOT: As an intermediate product of the trap-laser induced photoassociation process, a fraction of molecules exist in the first excited state at any given time. These molecules can be excited by the 12 815 cm\(^{-1}\) band to the 5s5d-asymptote and be ionized afterwards.

The energy potential curves of the 5p5p-asymptote (not depicted in Fig. 4) are situated below the 5s5d-asymptote and can, in principle, be reached by frequencies enclosed in the spectrum of the laser and also by two frequency bands of the optimized pulse. This excitation would start in the 5s5s states and evolve via the 5s5p to the 5p5p states by two photons of 12 815 cm\(^{-1}\) (780.4 nm). They are not involved in the ionization process since the ion yield vanishes if only the frequency components around 12 883 cm\(^{-1}\) (776.2 nm) are blocked.

Excluding these electronic states from the ionization process indicates that the molecules are very long-range bound molecules at the ionization step. At long binding distances the transitions follow the atomic selection rules. Regarding these, the transition from 5p5p to 5s+Rb\(^+\) cannot be carried out by a single photon. With a two or three photon transition the ionization could be accomplished, but due to the absence of resonance states it is too unlikely to be considered. In the case of wave packets being close to the inner turning point of the energy potential the selection rules are purely molecular. This would allow the direct ionization from states of the 5p5p-asymptote. Thus the states corresponding to the 5s5d-asymptote would not necessarily be preferred to the ones of the 5p5p-asymptote.

Another process in this experiment might be a free-bound transition in the first or second excitation step. This process of photoassociation cannot be distinguished from the excitation of molecules, which are already present in the trap.

The ionization step can be achieved by either the frequencies of 12 883 cm\(^{-1}\) (776.2 nm) or 12 815 cm\(^{-1}\) (780.4 nm) and leads to the \(\Sigma_g^+\) ground state of the ion. Other excited ionic states cannot be addressed with the provided energies of the optimized pulse. Due to the presence of repulsive potential energy curves and the considerable excess energy a high degree of fragmentation may occur in the ionic state.

VI. CONCLUSION AND OUTLOOK

In summary, we successfully optimized the ionization process of the rubidium dimer in a closed feedback loop optimal control experiment in a MOT. The observed two frequency bands were verified and analyzed by a second experiment. The electronic transitions were determined to belong to the 5s5s→5s5p\(_{3/2}\) and 5s5p\(_{3/2}\)→5s5d transitions. Furthermore it is revealed that the molecules are very long bound. The presented experiment shows an application of optimal control on ultracold atomic and molecular ensembles. This is a further step towards controlling reactions in a MOT or to transfer molecules to the vibrational ground state by complex shaped pulses.

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