

Coherent Control of Ultracold Molecule Dynamics in a Magneto-Optical Trap by Use of Chirped Femtosecond Laser Pulses

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We have studied the effects of chirped femtosecond laser pulses on the formation of ultracold molecules in a Rb magneto-optical trap. We have found that application of chirped femtosecond pulses suppressed the formation of $^{85}\text{Rb}_2$ and $^{87}\text{Rb}_2$ $a^3\Sigma_u^+$ molecules in contrast to comparable nonchirped pulses, cw illumination, and background formation rates. Variation of the amount of chirp indicated that this suppression is coherent in nature, suggesting that coherent control is likely to be useful for manipulating the dynamics of ultracold quantum molecular gases.

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Achieving control of the dynamics of quantum systems has been a long-standing goal of physics and chemistry [1,2]. Rapid advances in the manipulation of laser-matter interactions to obtain desired outcomes by means of tailored optical fields have been enabled by the development of ultrafast femtosecond optical sources and pulse-shaping techniques. In particular, recent successes include exciting *a priori* specified quantum states in atoms [3] and molecules [4], as well as in selectively cleaving chemical bonds in complex molecules [5]. Simultaneously, a very different thrust has been extending the regime of the ultracold ($T \leq 1$ mK) to simple molecular complexes [6]. Robust samples of trapped ultracold molecules are expected to facilitate significant advances in molecular spectroscopy, collision studies, and perhaps quantum computation [7,8].

The difficulty in generalizing laser cooling techniques to molecules has stimulated exploration of alternative approaches to producing ultracold molecules. One approach, using magnetic Feshbach resonances, has led to the observation of molecular Bose-Einstein condensates [9–11]. The study of other routes to ultracold molecule formation has also been a topic of intense activity [12]. An extremely successful optical approach has been to photoassociate molecules from ultracold atoms [13]. In this method, a sample of ultracold atoms is irradiated by a cw laser tuned to excite free atoms to weakly bound excited states. Stable ground-state molecules may then form by spontaneous emission, provided the Franck-Condon overlap factors are favorable. Ultracold molecules have been observed using this technique for a variety of homonuclear [14–17] and heteronuclear [18–21] alkali metal species.

Recent proposals have suggested controlling the interactions between ultracold atoms with tailored picosecond optical fields [22–25]. The possibility of improved formation of ultracold molecules using femtosecond coherent control techniques is therefore enticing. When a pulse length is < 100 ps, the light-matter interaction is entirely coherent, and therefore the details of the temporal shape of the pulse electric field may effect the interaction signifi-

cantly. This Letter describes the first experiments designed to enhance the molecule formation process in a magneto-optical trap (MOT) [26] with chirped femtosecond pulses. A schematic of the experiments is shown in Fig. 1. The broad bandwidth of femtosecond pulses permits excitation over a longer range of internuclear distances than picosecond pulses, thus addressing a larger number of atom pairs. Introducing a positively chirped molecular “ π ” pulse allows wave packet dynamics to strongly suppress detrimental population cycling [27]. This method also provides a route to optimal control of ultracold molecule formation [28]. Recent experiments using picosecond [29] and chirped nanosecond [30] pulses for photoassociation

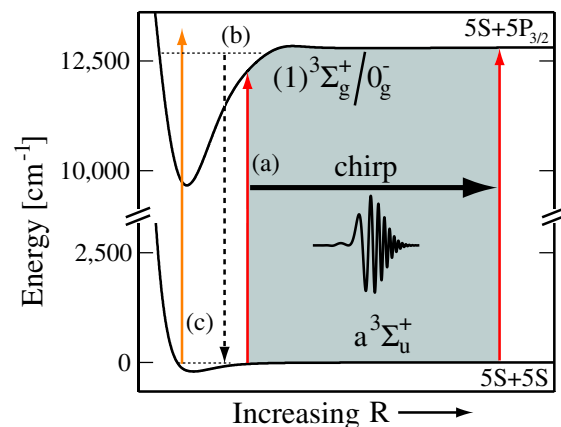


FIG. 1 (color online). Schematic of the chirped fs-pulse photoassociation experiment. The potential energy curves of Rb_2 involved in the design of the experiment are shown. (a) Ultracold atoms are excited over a range of internuclear separations by a chirped fs pulse to bound states below the $5S + 5P_{3/2}$ dissociation limit. The time-dependent frequency sweep ensures the unlikelihood of population cycling back to the ground state. (b) Excited molecules undergo spontaneous decay, some forming stable ground-state molecules. (c) These molecules are detected by resonance-enhanced two-photon ionization and TOF ion-mass spectroscopy.

(PA) provide encouragement for this approach. However, as we show in this Letter, the dynamics of molecule formation using femtosecond pulses appears to be significantly different from those predicted for picosecond pulses [23,24]. Here we report a suppression of molecule formation rather than an enhancement; nevertheless, the mechanism responsible for this effect appears to be coherent.

Femtosecond (fs) chirped-pulse PA was undertaken in an Rb MOT because its trapping transition (780.27 nm) is near the peak of the gain profile for Ti:sapphire (800 nm). Each isotope of Rb could be trapped by adjustment of the MOT trapping and repumping laser frequencies. Typical traps contained 2×10^7 atoms in a spheroidal cloud of diameter 0.7 mm, yielding a peak density on the order of 10^{10} cm^{-3} . The temperature of the atoms in the MOT was measured to be 170 μK using the release and recapture method [31].

Two different PA lasers were employed to study the effects of light on the molecule formation process in the MOT. The first was a cw free-running diode laser (Sanyo DL7140-201) with an output power of 56 mW, temperature tuned between 700–785 nm. The second laser was a commercial femtosecond-pulse oscillator (Spectra-Physics Mai Tai) whose carrier wavelength was tunable between 750–850 nm with output power ≤ 800 mW over this range. This laser had a repetition rate of 80 MHz and produced Gaussian-envelope transform-limited pulses of temporal intensity full-width at half-maximum (FWHM) < 100 fs, corresponding to a spectral bandwidth FWHM $> 150 \text{ cm}^{-1}$ for the available carrier wavelength range. Its output spectrum was monitored with a spectrometer and its field profile was regularly characterized using spectral interferometry for direct electric field reconstruction (SPIDER) [32].

Molecules formed in the MOT were detected via time-of-flight (TOF) ion-mass spectroscopy using a channel electron multiplier. A narrow-bandwidth (< 3 GHz) tunable pulsed dye laser (9 ns pulse duration, 50 Hz repetition rate) ionized the MOT cloud with pulses of energy 500 μJ . The PA and ionization lasers were merged onto the same beam path with a dichroic beam splitter and focused at the MOT cloud, ensuring that the sample that we ionized was the same as that affected by our PA lasers.

We chose to detect ground triplet state $a^3\Sigma_u^+$ Rb_2 molecules, which have been observed to form spontaneously in the MOT through one or both of two channels: PA stimulated by the MOT lasers themselves, and three-body recombination [16,33]. The ionization wavelength was tuned to 602.7 nm, which ionizes ground triplet state molecules via a resonant two-photon excitation ($X^2\Sigma_g \leftarrow 2^3\Pi_g \leftarrow a^3\Sigma_u^+$). To ensure that the detected molecules were in their electronic ground state, the MOT and PA lasers were shut off for at least 1 μs before the arrival of the ionization laser pulse to allow the atoms and molecules in the MOT to decay to their ground states. For a ^{85}Rb MOT the back-

ground detection rate of Rb_2^+ ions was typically 2–3 ions per pulse, while for a ^{87}Rb MOT the rate was significantly lower (0.3–0.8 ions per pulse).

A typical experiment involved applying one of the PA lasers to the MOT, applying the ionizing laser, and accumulating ion counts for a set number of ionization laser shots N_{shots} . A background scan with the PA laser blocked was then immediately acquired for another N_{shots} to determine the relative effect of the PA laser.

The cw PA laser was focused on the MOT with an intensity of around 10^3 W cm^{-2} . For an ^{87}Rb MOT, the molecular signal was increased for detunings of a few cm^{-1} below the trapping transition, with significant enhancement (by a factor of 1.5–2) at frequencies corresponding to resonant transitions to excited molecular vibrational states. For ^{85}Rb , the molecular signal was unaffected at resonant transition frequencies, and generally reduced below the background level by 50% at nonresonant frequencies. These results confirm those of Ref. [16], where the quenching phenomenon was interpreted as a coupling of molecules that form spontaneously in the ^{85}Rb MOT to excited dissociative states.

In order to promote efficient photoassociative excitations near the $D2$ line by the fs pulses, care was taken to select a pulse spectrum with power concentrated in the region just below the $5S + 5P_{3/2}$ dissociation limit (780 nm). The carrier wavelength of the fs laser was tuned to 783 nm. A spectral filter was used to ensure that the broad spectrum of the fs pulses did not contain significant power on the blue side of the $D2$ line (which would lead to direct excitation of atoms to repulsive states lying above the $5S + 5P_{3/2}$ dissociation limit). The fs-pulse power was attenuated to ≤ 0.3 nJ per pulse, corresponding to focused peak intensities on the order of 10^7 W cm^{-2} . Since this intensity is many orders of magnitude above the Rb $D2$ atomic transition saturation intensity (6 mW cm^{-2}), the light-matter interaction is beyond the perturbative regime.

Application of the spectrally filtered fs-pulse PA laser to ^{85}Rb and ^{87}Rb MOTs generally caused a strong reduction in the molecular ion signal as compared to background. In the case of a ^{85}Rb MOT, the quenching observed due to the fs PA laser was stronger than that induced by the cw PA laser. In the case of a ^{87}Rb MOT, contrary to the augmentative effect observed when the cw PA laser was applied, $\sim 50\%$ fewer molecular ions with respect to background were observed upon application of the fs laser under similar experimental conditions. $^{85}\text{Rb}_2^+$ ionization spectra for 600–610 nm recorded for the cases of no PA laser, cw PA laser, and fs PA laser had similar structure. Variation of the average power of the fs PA laser revealed evidence of an exponential-decay dependence of molecular ion yield on pulse power (see Fig. 2).

We further explored the effects of fs pulses on the molecular formation rate by varying the spectral chirp of the pulses. The 100 fs FWHM pulses were chirped by

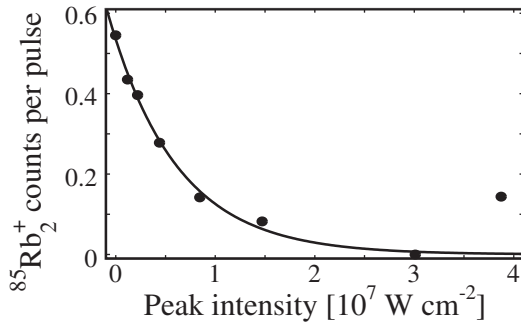


FIG. 2. $^{85}\text{Rb}_2^+$ counts per ionization pulse vs peak intensity of the applied fs pulses (filled circles). Each data point represents the accumulated result of an identical number of ionization laser shots. An exponential-decay fit of the data is shown (solid line).

passing the beam 13 times through a 5.0 in-length plane-parallel dispersive glass block. The chirped pulses had an estimated temporal intensity FWHM of 5.8 ps. To test whether the quenching effect involved resonant excitation of ground-state atoms by the fs laser, we also tuned its carrier wavelength to 850 nm (as far to the red as experimentally feasible) and removed the spectral filter. A spectrometer was used to confirm that the power spectrum of the chirped pulses did not differ substantially from that of nonchirped pulses.

Because of run-to-run drift of the MOT conditions and the significant time required to realign the setup when removing and reinserting the glass block into the PA laser beam path, we implemented a “real-time” differential measurement to compare the effects of chirped and nonchirped fs pulses. A schematic diagram of the experimental setup appears in Fig. 3. A broadband 50:50 beam splitter was used to split the fs laser beam into two identical components. The dispersive glass block was inserted into the path of one of these beams (hereafter the “chirped beam”). The second, transform-limited, beam (the “reference beam”) was attenuated with absorptive neutral density filters so that each beam had the same average power, to $\pm 1\%$ accuracy. An optical chopper wheel was aligned such that at any given instant one beam was blocked and the other was allowed to pass. Downstream from the chopper, the two beams were merged onto the same beam path using a second 50:50 broadband beam splitter. From this point forward, the two beams were aligned and focused to the MOT position as usual.

The experiment timing was set up so that for a single rotation of the chopper wheel, one chirped-pulse experiment and one reference pulse experiment occurred in sequence (see Fig. 3). The TOF data acquisition was performed in “toggle” mode: TOF traces acquired for chirped-pulse cases were added to the TOF histogram and traces acquired for reference pulse cases were subtracted from the histogram, yielding a net difference TOF histogram. A baseline differential measurement for the

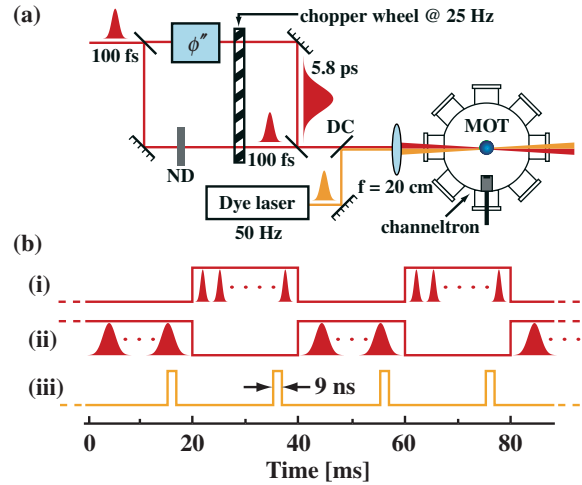


FIG. 3 (color online). (a) Schematic of the experimental setup. The fs PA beam was split by a 50:50 beam splitter; one beam was chirped by the dispersive glass block (ϕ''), and the other (reference) beam was power balanced using neutral density filters (ND). A chopper wheel synchronized with the detection apparatus allowed only one of the two beams to pass at a given instant. The two beams were merged at a second 50:50 beam splitter and focused to the MOT. The dye laser was merged onto the same beam path with a dichroic beam splitter (DC). (b) Reference and chirp beam detection timing. Alternating 80 MHz trains of reference (b-i) and chirped (b-ii) pulses were applied to the MOT. One dye laser ionization pulse (b-iii) fired for each train of pulses. Not shown here is the extinction of each pulse train for $1 \mu\text{s}$ prior to the firing of the dye pulse.

case with the dispersive glass block removed in Fig. 3 revealed a difference between two identical beams of fewer than 0.04 Rb_2^+ ions detected per ionization pulse.

The results of an experiment comparing the effects of a pulse train of 5.8 ps pulses to the effects of a pulse train of 100 fs pulses are shown in Fig. 4. The differential TOF trace reveals no discernible difference in the number of atomic ions, and a sharp negative feature in the vicinity of the arrival time of $^{85}\text{Rb}_2^+$ ions. The magnitude of this feature exceeded the baseline experiment Rb_2^+ detection rate by 400%. The negative feature in Fig. 4 signifies that appreciably more Rb_2^+ ions were accumulated after application of the transform-limited reference pulses. Thus, the chirped pulses quenched the Rb_2^+ ion signal more than the reference pulses. In the coherent regime the pulse area [$\propto \int E(t) dt$], not the pulse energy [$\propto \int |E(t)|^2 dt$], determines the nature of the light-matter interaction. The chirped and reference pulses were designed to have equivalent energies and different pulse areas. Direct comparison of these dissimilar pulses thus revealed that the quenching effect depended on the phase of the applied optical field. The quenching tended to increase with increasing chirp magnitude, although this trend was not monotonic as some chirp values were significantly more effective than others. These results provide evidence that the quenching effect is

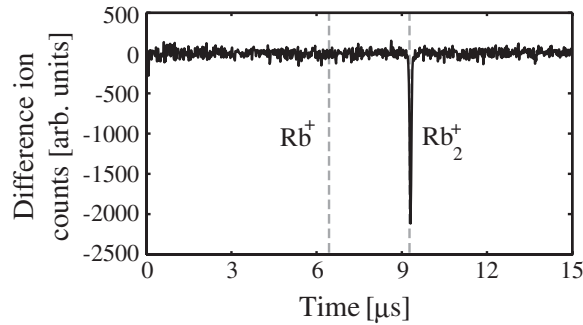


FIG. 4. Difference (chirped case—reference case) TOF histogram comparing the effects of chirped and transform-limited reference pulses. The vertical dashed lines indicate the approximate arrival times of ^{85}Rb atomic and molecular ions at the channeltron. There is no difference in the atomic ion signal between the two cases. A significant negative feature appears at the molecular ion arrival time, indicating that more Rb_2^+ ions were detected after applying the reference pulses than the chirped pulses.

a coherent phenomenon in which molecular quantum state dynamics play some role.

The exponential-decay dependence of the molecular formation rate on applied fs-pulse peak intensity is consistent with a single-photon excitation loss from either the detected $a^3\Sigma_u^+$ or the excited $1^3\Sigma_g^+/0_g^-$ states. There are several coherent excitation pathways for 850 nm light that could be responsible for a reduction in the number of detected $a^3\Sigma_u^+$ molecules. One is MOT-laser PA followed by excitation of excited molecules in the $1^3\Sigma_g^+/0_g^-$ states to bound states in potentials that dissociate to $5P + 5P$ ($6^3\Sigma_u^+$, $7^3\Sigma_u^+$, and $4^3\Pi_u$)—all of which are “dark” to our detection scheme—or to dissociative states lying above the $5S + 6P$ dissociation limit. Another possibility is that the population is excited from the ground state $a^3\Sigma_u^+$ onto the C_3R^{-3} potential of the $1^3\Sigma_g^+$ state. The excited state spatial wave function may have enhanced Franck-Condon overlaps with either continuum states lying above the $5S + 5S$ dissociation limit or undetected $a^3\Sigma_u^+$ bound states, thus resulting in a decrease in the number of detected $a^3\Sigma_u^+$ molecules [34].

In summary, we observed that application of fs pulses designed to photoassociate ultracold atoms resulted in a decrease, rather than an increase, in the formation of stable ultracold $a^3\Sigma_u^+$ molecules. Related work has recently been undertaken which provides evidence that the shape of the pulse spectrum has an effect on the quenching of the molecular formation rate [35]. We have shown here that phase shaping of the applied fs pulses can be used to control the quenching rate. This result provides evidence that the quenching is a coherent process, although we are unable to distinguish the specific excitation pathway stimulated by the fs pulses. Further experimental and theoretical study of fs pulses for optimal control of photoassociative

ultracold molecule formation is warranted, particularly for the creation of stable singlet molecules whose deeper $X^1\Sigma_g^+$ potential may be better suited to the broad inherent bandwidth of fs pulses.

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