Coherent control of ultrafast bond making and subsequent molecular dynamics: demonstration of final-state branching ratio control

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Abstract
Quantum coherent control of ultrafast bond making and the subsequent molecular dynamics is crucial for the realization of a new photochemistry, where a shaped laser field is actively driving the chemical system in a coherent way from the thermal initial state of the reactants to the final state of the desired products. We demonstrate here coherent control over the relative yields of Mg2 molecules that are generated via photoassociation and subsequently photodriven into different groups of final states. The strong-field process involves non-resonant multiphoton femtosecond photoassociation of a pair of thermally hot magnesium atoms into a bound Mg2 molecule and subsequent molecular dynamics on electronically excited states. The branching-ratio control is achieved with linearly chirped laser pulses, utilizing the different chirp dependence that various groups of final molecular states display for their post-pulse population. Our joint experimental and theoretical study establishes the feasibility of high degree coherent control over quantum molecular dynamics that is initiated by femtosecond photoassociation of thermal atoms.

Keywords: coherent control, femtosecond coherent control, quantum coherent control, coherent control of bond making, ultrafast bond making

A driving force for the field of quantum coherent control has been the idea of generating a new type of photochemistry [1–4] in which a shaped laser field actively drives the irradiated quantum system in a coherent way from the initial state of the reactants to the final state of the desired products. This is conceptually different from the common photochemistry approach, where the reactants are individually photo-excited and then react without further interaction with external fields. The key tool for the coherent control of binary reactions are shaped femtosecond laser pulses [5]. Typically, coherent control of a binary reaction proceeds in several steps. First, ultrafast coherent bond making (i.e. free-to-bound femtosecond photoassociation) is photo-induced between the free colliding reactants, and an excited bound molecule is generated. Then, subsequent photo-control over the molecular dynamics directs...
the system into target intermediate states. Last, these inter-
mediate states serve as a platform for further coherent photo-
control in order to selectively break bonds (i.e. femtosecond
photodissociation) for generating the desired products.

Despite the significance, there is still no realization of this
full scheme of binary reaction control from reactants to
products. A major reason is the lack of successful coherent
control of ultrafast bond making and the subsequent molec-
ular dynamics. For binary gas phase photo-reactions where
the reaction mechanism is most easily unraveled, such con-
tral has been demonstrated so far only in a few studies we
have conducted [6–9]. Control of photoassociation was also
demonstrated at ultralow temperatures with a timescale five
orders of magnitude longer [10–13]. Quantum control of bond
formation was also observed in laser-induced catalytic sur-
face reactions [14, 15] without, however, full insight into the
reaction mechanism. The key element is distillation of an
entangled pair from a thermal ensemble. In strong contrast,
many studies have successfully demonstrated photodissocia-
tion control of bound molecules, controlling both the total
yield of fragments as well as their branching ratio to different
channels [16–25].

The strong-field process in our previous photoassociation
investigations [6–9] involves non-resonant multiphoton
femtosecond photoassociation of a pair of thermally hot mag-
nesium atoms into a bound excited Mg$_2$ molecule and subse-
quent molecular dynamics on electronically excited states.
At the first stage, we have demonstrated the generation of purity
and rovibrational coherence in the ensemble of photoassoci-
ated Mg$_2$ molecules via the mechanism of Franck–Condon
filtering [6, 7]. Then, we have utilized the generated purity
and coherence to demonstrate coherent control over the result-
ing yield of molecules populating a single group of final
states [8, 9]. The molecular yield was enhanced with linearly-
chirped femtosecond pulses having a positive chirp, reach-
ing a maximum with a specific optimal chirp value, as well
as with positively chirped pulses split into sub-pulses with a
temporal structure that fits the photo-induced coherent vibra-
tional dynamics. The different excitation and control mech-
nisms have been qualitatively explained by our ab initio
calculations.

In view of controlling the full scheme of a binary
reaction from reactants to products, the main limitation of our
previously demonstrated bond-making control has been the
control target, i.e. the overall molecular yield [6–9]. Here, we extend our approach to coherently con-
trolling the relative yields of molecules that are gener-
ated via photoassociation and subsequently photo-driven into
multiple groups of final states. The investigated ultrafast
strong-field process is, as before, photoassociation of ther-
mal magnesium atoms followed by excited-state dynamics of
the generated Mg$_2$ molecules. The demonstrated branching-
ratio control is achieved with linearly-chirped pulses, util-
izing the different chirp dependence that different groups
of final molecular states display for their post-pulse popu-
lation. Our results establish the feasibility of high degree
coherent control over quantum molecular dynamics that
is initiated by femtosecond photoassociation of thermal
atoms.

Figure 1 shows the potential energy curves for the magne-
sium dimer [7–9] together with the ultrafast excitation scheme.
The starting point is an ensemble of magnesium atoms at a
temperature of 1000 K that thermally populates scattering
states above the van-der-Waals ground electronic state X$^1\Sigma_g^+$
of Mg$_2$, indicated by gray shading in figure 1. The ensemble
is irradiated with shaped femtosecond laser pulses having a
central wavelength of 840 nm, a transform-limited (TL) dura-
tion of 70 fs, linear polarization, and an energy that corre-
sponds to a TL peak intensity of 7.2 × 10$^{12}$ W cm$^{-2}$. The pulse
photo-associates pairs of magnesium atoms and generates Mg$_2$
molecules via a free-to-bound non-resonant two-photon trans-
ition from X$^1\Sigma_g^+$ scattering states to rovibrational states of
the $^1\Pi_u$ state. The corresponding Franck–Condon window
is located at short internuclear distances. Then, the pulse further
induces subsequent molecular dynamics on the $^1\Pi_u$ and higher
electronically excited states, resulting in a post-pulse popula-
tion that spans a manifold of final molecular states. Our present
objective is to control the population branching ratio among
final states of the photoassociated Mg$_2$ molecules that
cover an extended energy band located below and above the
$^1\Pi_u$ asymptote, indicated also by gray shading in figure 1.
These states belong to the A$^1\Sigma_u^+$, (1)$^1\Pi_u$, (2)$^1\Pi_u$, or (2)$^1\Sigma_u^+$
electronically excited states, which are all dipole-coupled to
the $^1\Pi_u$ and X$^1\Sigma_g^+$ states. The $^1\Pi_u$, A$^1\Sigma_u^+$ and (1)$^1\Pi_u$
states share the same asymptote of Mg(1$^3P$) + Mg(1$^3S$), having with
the ground-state asymptote Mg(1$^3S$) + Mg(1$^3S$) an atomic transi-
tion energy of 35 051 cm$^{-1}$ corresponding to a measured
wavelength of $\lambda_0 = 285.2$ nm. For the control, we employ
linearly-chirped femtosecond laser pulses, all having the same
spectrum but different spectral phase. Their spectral phase is
of the form $\Phi(\omega) = \frac{\pi}{2} k (\omega - \omega_0)^2$, where $\omega$ is a given frequen-
cy, $\omega_0$ the central frequency of the pulse spectrum (corresponding
here to a wavelength of 840 nm) and $k$ the linear chirp parame-
ter. The latter ($k$) is the control variable. The unchirped, or TL,
pulse corresponds to $k = 0$.

Experimentally, magnesium vapor with a pressure of about
5 Torr is held in a heated static cell at 1000 K with Ar buffer
gas, and the sample is irradiated at 1 kHz repetition rate with
the intense shaped femtosecond pulses described above. They
are phase shaped using a liquid-crystal spatial light modula-
tor [5]. The post-pulse population of the final molecular states
is probed by detecting and spectrally resolving the ultraviolet
(UV) radiation emitted in their spontaneous radiative decay to
the ground electronic state X$^1\Sigma_g^+$. The spectral range of the
detected UV emission is $\lambda_{uv,s} = 281.5$ nm to $\lambda_{uv,l} = 287.5$ nm
(34 783–35 524 cm$^{-1}$). In the setup, the UV radiation emitted
toward the laser-beam entrance to the cell is collected at a small
angle from the laser-beam axis using an appropriate optical
setup, spectrally resolved by a spectrometer with 0.1 nm reso-
lation, and then intensity-measured using a time-gated camera
system with a post-pulse gate of 20 ns. The spectral range
and resolution of the UV emission are the major differences to
our earlier studies [6–9] which measured only the total UV emis-
sion intensity integrated over the range of wavelengths longer
than $\lambda_0$. 

Figure 1. Potential energy curves of Mg₂ and ultrafast excitation scheme corresponding to an intense phase-shaped near-infrared linearly-polarized femtosecond pulse. The strong-field ultrafast excitation involves bond making (free-to-bound photoassociation) and subsequent molecular dynamics on the excited electronic states. The observable spectrum is the sum of the post-pulse emission spectra resulting from the excited post-pulse population which decays by UV emission.

The potential energy curves are plotted for the rotational quantum number $J = 70$, which corresponds roughly to the maximum of the initial thermal rotational population. The gray shades indicate the full map, figure 3(a) presents examples of the measured emission spectrum an individual spectrum $[I^{(f)}_{\text{em}}(\lambda_{uv}, k)]$ with a given state-specific shape $[S^{(f)}_{\text{em}}(\lambda_{uv})]$. The total intensity $[I^{(f)}_{\text{em, tot}}(k)]$ for each state is proportional to the number of Mg₂ molecules populating that state $[N^{(f)}_{\text{mol}}(k)]$. The corresponding proportionality constant $[\alpha^{(f)}_{\text{em}}]$ depends on the different radiative channels that the state can spontaneously decay through, and stands for the intensity fraction of the state’s emission at the UV spectral range detected here out of the full emission from that state. Hence, a change in the shape of the total measured spectrum $[S^{(f)}_{\text{em}}(\lambda_{uv}, k)]$ can result only from a corresponding change in the relative individual contributions of the different final states to the total spectrum. Such a change can occur due to a change in the relative population $[p^{(f)}(k) = N^{(f)}_{\text{mol}}(k)/\sum_f N^{(f)}_{\text{mol}}(k)]$ of multiple final states. In other words, the observed chirp-dependent changes of $S^{(f)}_{\text{em}}(\lambda_{uv}, k)$ directly imply corresponding chirp-dependent changes in the relative yields of photoassociated molecules populating the different final molecular states, i.e., coherent chirp control over the corresponding yield branching ratio.

The associated quantitative analysis is as follows. After the excitation with a pulse of chirp $k$, the emission spectrum from a final state $f$ is given by $I^{(f)}_{\text{em}}(\lambda_{uv}, k) = \alpha^{(f)}_{\text{em}} \cdot N^{(f)}_{\text{mol}}(k) \cdot S^{(f)}_{\text{em}}(\lambda_{uv})$, such that the state-specific shape $S^{(f)}_{\text{em}}(\lambda_{uv})$ satisfies $\int_{\lambda_{uv}} S^{(f)}_{\text{em}}(\lambda_{uv}) d\lambda_{uv} = 1$. The chirp-dependent shape of the total spectrum emitted from a group of states is then given by $I^{(f)}_{\text{em, tot}}(\lambda_{uv}, k) = \sum_f W^{(f)}(k) \cdot S^{(f)}_{\text{em}}(\lambda_{uv})$ where $W^{(f)}(k) = \frac{\alpha^{(f)}_{\text{em}} \cdot p^{(f)}(k)}{\sum_f \alpha^{(f)}_{\text{em}} \cdot p^{(f)}(k)}$. Both $\alpha^{(f)}_{\text{em}}$ and $S^{(f)}_{\text{em}}(\lambda_{uv})$ do not depend on the chirp $k$, while $p^{(f)}(k)$ might have such a dependence. Thus, indeed, the changes observed for $S^{(f)}_{\text{em}}(\lambda_{uv}, k)$ when tuning the chirp $k$ necessarily reflect changes in the relative population $p^{(f)}(k)$ of multiple final states.

The chirp dependence of the measured emission spectrum seen in figures 2 and 3 includes several prominent characteristics. In terms of the total spectral intensity $I^{(f)}_{\text{em, tot}}(k)$, the positively-chirped pulses enhance it with respect to the TL pulse, while the negatively-chirped pulses attenuate it. In terms of the spectral shape $S^{(f)}_{\text{em}}(\lambda_{uv}, k)$, from chirp zero up to a positive chirp $k$ of 0.006–0.007 ps², it exhibits only weak chirp dependence and its dominant part is the longer-wavelength part of $\lambda_{uv} > \lambda_a$. Then, as the chirp $k$ increases to larger positive values, the spectral shape shifts to shorter wavelengths, with an enhancement of its shorter-wavelength part and an attenuation of its longer-wavelength part. In terms of the complementary characteristic of the emission intensity $I^{(f)}_{\text{em}}(k; \lambda_{uv})$ at a given wavelength $\lambda_{uv}$, for all the detected wavelengths, it is enhanced with positively-chirped pulses over an extended range of positive chirps, and has a maximum at an emission wavelength-dependent positive chirp $k^{(f)}_{\text{max}}(\lambda_{uv})$. The value of $k^{(f)}_{\text{max}}(\lambda_{uv})$ stays unchanged (as 0.006 ps²) for $\lambda_a < \lambda_{uv} < \lambda_a W$, and then continuously increases as $\lambda_{uv}$ continuously decreases from $\lambda_a$ down to $\lambda_a W$.

Our interpretation for the mechanism facilitating the branching-ratio control is built upon the dynamics we have previously [8, 9] identified to be induced by the positively chirped pulses. Subsequent to the $^1 \Sigma_u^+ \rightarrow ^1 \Pi_g$ free-to-bound two-photon transition, the strong-field dynamics taking place on the $^1 \Pi_g$ state involves multiple resonant Raman transitions...
Figure 2. Experimentally detected UV emission intensity as a function of emission wavelength $\lambda_{uv}$ and chirp parameter $k$ of the linearly chirped femtosecond pulses. The intensity values are color coded, and the black dashed line indicates the wavelength $\lambda_a$ corresponding to emission at the $^1\Pi_g$ atomic asymptote.

(via higher electronically excited states) that lead to a vibrational de-excitation, i.e., a population transfer into vibrational levels that are lower than the levels accessed by the photoassociative two-photon transition. Then, final perturbative one-photon absorption projects the vibrationally de-excited $^1\Pi_g$ population onto the final molecular states that emit UV light when decaying to the $X^1\Sigma_g^+$ state. The higher the energetic location of the emitting states, the higher is the energetic location of the $^1\Pi_g$ vibrational region from which they are effectively accessed by the final projection. Following their different locations, the population transfer to distinct vibrational regions is induced by a different number of Raman transitions. So, with a given pulse energy, each positively chirped pulse, due to its unique temporal intensity profile (i.e. a Gaussian of unique width), generates a different distribution of relative population among the various $^1\Pi_g$ vibrational regions and, thus, also among the various final emitting states. This, in turn, results in a different shape for the total emission spectrum $S_{\text{em}}(\lambda_{uv}; k)$. This is the control mechanism of the present chirp-dependent branching ratio among the different final states.

Furthermore, our previous results [8] also indicate that the chirp $k_{\text{max}}$ which maximizes the population transfer into a given $^1\Pi_g$ vibrational region, corresponds to the positively-chirped pulse having the longest temporal segment with instantaneous intensities $J_{\text{pulse}}(t)$ that are all above a certain threshold intensity $J_{\text{th}}^{(\text{vib. region})}$. The latter is associated with the vibrational region. Increasing the chirp $k$ from zero up to this $k_{\text{max}}^{(\text{vib. region})}$ lengthens this segment up to a maximal duration, and a further increase in $k$ leads to its shortening (eventually down to zero duration). For a given pulse energy, $k_{\text{max}}^{(\text{vib. region})}$ is inversely proportional to $J_{\text{th}}^{(\text{vib. region})}$. In other words, in order to increase the strong-field de-excitation efficiency into a specific $^1\Pi_g$ vibrational region, it is beneficial to temporally stretch the positively-chirped pulse as much as possible up to a certain limit (that depends on the vibrational-region’s location). This fits the strong-field non-perturbative nature of the de-excitation dynamics. Since an efficient population transfer to a higher $^1\Pi_g$ vibrational region requires a smaller number of efficient Raman transitions, the corresponding threshold intensity $J_{\text{th}}^{(\text{vib. region})}$ is smaller and the corresponding $k_{\text{max}}^{(\text{vib. region})}$ is larger. As emitting states of higher excitation energy are linked with a higher vibrational region, their population and emitted radiation will thus be maximally enhanced with a larger chirp value as compared to emitting states of lower excitation energy. Hence, the present experimental chirp dependence of the total spectral shape $S_{\text{em}}(\lambda_{uv}; k)$ and the emission wavelength-dependent optimal chirp $k_{\text{max}}(\lambda_{uv})$ point toward the following characteristic for the states that dominantly contribute to the detected emission: the higher the energy of the emitting state, the shorter are the UV wavelengths at which its state-specific emission spectral shape $S_{\text{em}}^{(f)}(\lambda_{uv})$ is intensified.
intensity. See text for details.

obtained by normalizing the measured spectrum by its total at the corresponding chirp. For each emission spectrum, panel (a) each spectrum corresponds to a cut of the emission map of figure2

mental detected range of states below the Mg(1

tional levels of the two coupled 1

Examples of experimental UV emission spectra measured femtosecond pulses for different values of the chirp parameter k. Each spectrum corresponds to a cut of the emission map of figure 2 at the corresponding chirp. For each emission spectrum, panel (a) presents the measured spectrum and panel (b) the spectral shape obtained by normalizing the measured spectrum by its total intensity. See text for details.

Considering this characteristic in combination with the results of the theoretical emission analysis described next allows us to identify the dominant emitting states.

Using the potential curves presented in figure 1 and the corresponding electronic transition dipole moments [7–9], we have assigned the origin of the experimentally detected UV emission to different electronic states and vibrational levels. First, we employ energetic considerations to group the different states according to their emission wavelength. Second, analysis of the corresponding vibrationally-averaged transition dipole moments (VTDMs) with the electronic ground state X1Σg+ allows us to calculate the full spectrum of spontaneous emission from each state in these groups. In the experimentally detected range of λuv,x < λuv < λuv,L, four such distinct sets of states can be identified: (I) high-lying vibrational levels of the A1Σu+ state near the Mg(1P) + Mg(1S) asymptote, (II) vibrational levels of the two coupled 1Πu states below the Mg(1P) + Mg(1S) asymptote, (III) vibrational levels of the two coupled 1Πu states above the Mg(1P) + Mg(1S) asymptote but below the avoided crossing of the 1Πu’s potentials, and (IV) low-to-moderately high-lying vibrational levels of the (2)1Σu+ state. States from groups (I) and (II) contribute exclusively to the emission at wavelengths longer than λu. States from groups (III) and (IV) contribute both to the emission at wavelengths longer and shorter than λu. Even though it would be energetically possible for even higher-lying states to add to the detected emission window by decaying into vibrationally high-lying levels in the electronic ground state, the VTDMs for such transitions are effectively zero. Moreover, vibrational levels on the two coupled 1Πu states above the avoided crossing are subject to predissociation which is rapid compared to the spontaneous-emission timescale. This implies that their contribution to UV emission can be neglected, and group (III) only contains levels below the avoided crossing.

Figure 4 shows the group-specific UV emission spectra in the experimentally detected range, calculated for rotational quantum numbers J = 30, 70 and 120, with J = 70 corresponding roughly to the maximum of the initial thermal rotational population. The results are shown as grayscale-coded maps, with each map corresponding to a different group. For each vibrational level in a group, the map shows the relative emission intensity at each wavelength assuming the initial population of the level is 100%. A careful analysis of the calculated VTDMs and emission spectra for all rotational levels reveals that, for group (II), only very few or no vibrational levels contribute to the experimental emission window. For the example of J = 70 shown in figure 4, only a single contributing level remains. Furthermore, the group-specific emission intensities within the experimental emission window are larger for group (I) by two orders of magnitude as compared to all other groups. Due to the fact that group (I) only contributes to emission at wavelengths longer than λu, we conclude that the detected emission at this range is dominated by group (I), with several dozen vibrational levels showing large VTDM values and high emission intensities. In particular, the density of vibrational levels in group (I) is highest right below the asymptote of Mg(1P) + Mg(1S), which leads to an accumulation of the emission intensity directly at wavelengths slightly longer than λu. As seen in figure 4, the identification of group (I) as dominant in the emission spectral range of λu < λuv < λuv,L indeed fits the characteristic identified above for the dominant emitting states with a correlation between higher state energy and intensified emission at shorter wavelengths. Moreover, this identification of group (I) agrees with our results from previous chirp-dependent experiments and quantum dynamical calculations for the long-wavelength emission [8].

With regard to the emission at wavelengths shorter than λu, figure 4 shows that in this spectral range only group (III) follows the aforementioned characteristic of the dominant emitting states. Hence, we conclude that the detected emission at the spectral range of λuv,L < λuv < λu is dominated by the states of group (III). This is also consistent with the larger state-specific emission intensities seen in figure 4 for group (III) as compared to group (IV). Overall, we attribute the detected emission at λuv > λu primarily to high-lying vibrational levels of the A1Σu+ state located near the Mg(1P) +
Figure 4. State-specific UV emission spectra in the experimentally detected range calculated for the vibrational levels with rotational quantum number $J = 30$, $70$ and $120$ that belong to the four different groups of electronically excited states which may contribute to the detected emission. Each (grayscale-coded) map corresponds to a different group, and for each vibrational level in a group, the map shows the relative emission intensity at each wavelength assuming the initial population of the level is $100\%$. Note the different color bar scales for the four groups.

Mg($1S$) asymptote, whereas the detected emission at $\lambda_{uv} < \lambda_a$ predominantly appears to originate from vibrational levels of the two coupled $1\Pi_u$ states located above the Mg($1P$) + Mg($1S$) asymptote but below the avoided crossing.

In summary, coherent control of ultrafast bond making and subsequent molecular dynamics in experiment and theory has been extended here to demonstrating branching ratio control. The relative yields of molecules, which are coherently generated in thermal femtosecond photoassociation and subsequently photo-driven into different target states, are coherently controlled using linearly-chirped pulses of positive chirp. The control knob is the chirp of the pulse. The results are explained by calculations of the UV emission together with a model accounting for vibrational de-excitation that is photo-induced and controlled via strong-field chirped Raman transitions. Our demonstrated control is a crucial element for the realization of coherent control over the complete process of a binary photo-reaction. In the case of a reaction with several product channels, the branching ratio control to various target molecular states will be the first part of an extended scheme, in which the different target states will serve as intermediate platforms from which subsequent selective photo-control will be applied toward different product channels.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).
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