Laser Induced Selective Alignment of Water Spin Isomers

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We consider laser alignment of ortho and para spin isomers of water molecules by using strong and short off-resonance laser pulses. A single pulse is found to create a distinct transient alignment and antialignment of the isomeric species. We suggest selective alignment of one isomeric species (leaving the other species randomly aligned) by a pair of two laser pulses.

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I. INTRODUCTION

According to quantum mechanics, a molecule that contains two identical atoms whose nuclei have nonzero spins can exist in the form of spin isomers. In particular, a water molecule exists in one of the two spin isomers, ortho or para with parallel or antiparallel proton spins, respectively. The ortho-para conversion is highly improbable in the gas phase [1]. Water isomers separation (or their ratio measurement) can be of great importance in medicine, biology, chemistry, magnetic resonance imaging, etc. Measurement of the para to ortho ratio was used to estimate the formation temperature of molecules in space [2]. A method for mixture enrichment in one of the water isomers was reported using selective adsorption onto various surfaces [3], although some of these results were hardly reproducible. Recently Andreev et al. [4] discussed an expected difference in the response of the ortho and para forms of water to a non-uniform dc electric field near solid surfaces, which may potentially lead to isomers separation. We are currently exploring an alternative route to ortho and para separation based on selective field-free alignment and antialignment of water isomers by short nonresonant laser pulses (for a review on field-free alignment see [5]). By alignment we mean the angular localization of a molecular axis along the laser polarization direction, while antialignment assumes localization of the same molecular axis in the plane perpendicular to the laser polarization. Although previous experimental studies on dissociative fragmentation by relatively long laser pulses have not found alignment of water molecules [6], we show that a sizable alignment can be achieved in ultra-short regime. The selective alignment was recently experimentally demonstrated in a mixture of ortho and para modifications of linear molecules (\(^{15}\)N\(_2\) considered as an example) [7]. Here, for the first time to our knowledge, we consider selective alignment for bent tri-atomic molecules like water. The paper is organized as follows. We briefly discuss the classification of the states of the ortho and para water molecules in Sec. II. The interaction of the water molecules with the laser field is considered in Sec. III. The results are presented in Sec. IV and briefly summarized in Sec. V. In the Appendix, we give a detailed description of the rotational wavefunctions of the ortho and para water isomers.

II. CLASSIFICATION OF THE EIGENSTATES OF THE WATER MOLECULE

The water molecule wavefunction is given by:

\[
|\Psi\rangle = |\Psi_{\text{rot}}\rangle|\Psi_{\text{spin}}\rangle|\Psi_{\text{vib}}\rangle|\Psi_{\text{ele}}\rangle ,
\]

where \(|\Psi_{\text{rot}}\rangle\), \(|\Psi_{\text{spin}}\rangle\), \(|\Psi_{\text{vib}}\rangle\) and \(|\Psi_{\text{ele}}\rangle\) are rotational, nuclear spin, vibrational and electronic wavefunctions, respectively. The water molecule eigenstates are classified using the \(C_{2v}(M)\) Molecular Symmetry (MS) group [1], i.e. using the irreducible representations \(A_1\), \(A_2\), \(B_1\), \(B_2\). We provide the group operations as well as its character table in the Appendix. According to the Pauli Exclusion Principle, the total wavefunction should change its sign after permutation of the hydrogen nuclei. Since we consider \(|\Psi_{\text{vib}}\rangle\) and \(|\Psi_{\text{ele}}\rangle\) in the ground state (which is symmetric with respect to the \(C_{2v}(M)\) operations), only \(|\Psi_{\text{rot}}\rangle\) and \(|\Psi_{\text{spin}}\rangle\) are effected by the operations. The rotational part of the water molecule Hamiltonian is presented by the rigid rotor model:

\[
\hat{H} = \frac{\hat{J}_a^2}{2I_a} + \frac{\hat{J}_b^2}{2I_b} + \frac{\hat{J}_c^2}{2I_c} ,
\]

where \(\hat{J}\) is the angular momentum operator, \((a, b, c)\) are the molecule’s principal axes, and \(I_a, I_b, I_c\) are the corresponding moments of inertia \((I_a < I_b < I_c)\). Rotational states of \(A_1\) and \(A_2\) symmetries do not change sign as a result of permutation and belong to the para states (with anti-parallel spins), while the states of \(B_1\) and \(B_2\) symmetries correspond to the ortho states. Further information about the water molecule eigenstates classification is given in the Appendix.

III. INTERACTION OF THE WATER MOLECULES WITH LASER FIELD

We consider the alignment process based on interaction with short off-resonance laser pulses that induce molecular polarization, interact with it, and excite rotational wavepackets. The interaction of the water molecule (shown in Fig. 1) with the laser pulse is given by [8]:

\[
H_{\text{int}} = -\frac{1}{4} \sum_{\rho, \rho'} \varepsilon_{\rho}(t) \alpha_{\rho, \rho'} \varepsilon_{\rho'}(t) ,
\]

where \(\varepsilon\) is the electric field of the laser pulse.
where the $\rho$ index denotes the space-fixed Cartesian coordinates, $\varepsilon_{\rho}$ is the laser pulse envelope component, and $\alpha$ is the polarizability tensor.

For linearly polarized fields oriented in the laboratory $Z$ direction, the interaction term is reduced to:

$$
H_{\text{ind}} = \frac{\varepsilon^2(t)}{4} \left[ \alpha_{ab} \cos^2 \theta + \alpha_{cb} \sin^2 \theta \sin^2 \chi \right]
$$

where $\alpha_{ab} = \alpha_{aa} - \alpha_{bb}$, etc., $D_{mk}^J$ are the rotational matrices [10], and $\hat{R} \equiv (\theta, \phi, \chi)$.

In the case of a symmetric top molecule (i.e. when $\alpha_{xx} = \alpha_{yy} = \alpha_{zz}$ and $\alpha_{zz} = \alpha_{yy}$), the Hamiltonian is reduced to the well-known form: $H_{\text{ind}} = -\frac{1}{2} \varepsilon^2(t) \Delta \alpha \cos^2 \theta$, where $\Delta \alpha = \alpha_{\|} - \alpha_{\perp}$. In the case of water, the polarizability tensor components are: $\alpha_{aa} = 1.528 \AA^3$, $\alpha_{bb} = 1.468 \AA^3$ and $\alpha_{cc} = 1.415 \AA^3$ [11].

Given an initial rotational eigenstate of the asymmetric water molecule, we are interested in the field-free evolution of the rotational subsystem after an excitation by a laser pulse that is short compared to the typical rotational periods of the water molecule. Using the impulsive approximation, we consider only the contribution from the interaction term to the Schrödinger equation during the pulse:

$$
\{ \gamma_1 D_{00}^2 + \gamma_2 [D_{02}^2 + D_{0-2}^2] \} \Psi = \frac{i}{\hbar} \frac{d \Psi}{d t},
$$

where $\gamma_1$ and $\gamma_2$ are found from Eq. (4).

Solving Eq. (5), we obtain the relation between the states before and after the pulse:

$$
\Psi(t = 0^+) = \exp \left\{ \frac{1}{i} \left[ \beta_1 D_{00}^2 + \beta_2 [D_{02}^2 + D_{0-2}^2] \right] \right\} \Psi(t = 0^-),
$$

where $\beta_{1,2} = (1/\hbar) \int \gamma_{1,2} dt$.

In order to find $\Psi(t = 0^+)$, we introduce an artificial parameter $\xi$ that will be assigned the value $\xi = 1$ at the end of the calculations:

$$
\Psi_\xi = \exp \left\{ \frac{1}{i} \left[ \beta_1 D_{00}^2 + \beta_2 [D_{02}^2 + D_{0-2}^2] \right] \xi \right\} \Psi(t = 0^-).
$$

We express $\Psi_\xi$ in terms of the water molecule eigenstates:

$$
\Psi_\xi = \sum_{J, \tau} c_{J, \tau}^\xi | J, \tau, m \rangle,
$$

where $J$ is the angular momentum quantum number, and $m$ is the quantum number of the angular momentum projected on the laboratory $Z$ axis. For the chosen laser field
polarization, \( m \) is a conserved quantum number. This allows us to consider the excitation of the states with a different initial \( m \)-value separately. Index \( \tau \) (which is assigned the values \(-J,\ldots,+J\)) numerates various \( 2J + 1 \) eigenstates arranged from the lowest energy (\( \tau = -J \)) to the highest energy (\( \tau = +J \)).

Then we take a derivative of both sides of Eq. (7) with respect to \( \xi \), apply the bra \( \langle J', \tau', m \rangle \) from the left, and obtain a set of differential equations:

\[
\hat{c}_{J', \tau'} = \frac{1}{i} \sum_{J, \tau} c_{J, \tau}^{+} \{ \beta_{1} \langle J' \tau' m | D_{02}^{2} | J \tau m \rangle + \beta_{2} \langle J' \tau' m | D_{02}^{2} | J \tau m \rangle \}, \tag{9}
\]

where \( \hat{c} \equiv dc/d\xi \).

Since \( \Psi_{\xi=0} = \Psi(t = 0) \) and \( \Psi_{\xi=1} = \Psi(t = 0^{+}) \) (Eq. (9)), we solve numerically this set of equations from \( \xi = 0 \) to \( \xi = 1 \), and find \( \Psi(t = 0^{+}) \).

The post-pulse field-free propagation of the created rotational wave packet is calculated as

\[
\Psi(t) = \sum_{J, \tau} c_{J, \tau}^{+} \exp \left\{ -i \frac{E_{J'}}{\hbar} t \right\} |J, \tau, m\rangle, \tag{10}
\]

where \( E_{J'} \) is the eigenvalue corresponding to the eigenstate \( |J, \tau, m\rangle \). We calculate the matrix elements in Eq. (9) by expressing the water molecule eigenstates using the basis of the symmetric rotor: \( |J, k, m\rangle \) (\( J \) and \( m \) are defined as before, and \( k \) is the quantum number of the angular momentum projected on the rotor symmetry axis), and using the following relations:

\[
\langle Jkm|D_{02}^{2}|J'k'm\rangle = (\pm 1)^{k'-m} \times \sqrt{(2J+1)(2J'+1)} \begin{pmatrix} j & 2 & j' \hline m & 0 & -m \end{pmatrix} \begin{pmatrix} j & 2 & j' \hline k & s & -k' \end{pmatrix}. \tag{11}
\]

Here \( s = 0, \pm 2 \), and \( \begin{pmatrix} j_{1} & j_{2} & j_{3} \hline m_{1} & m_{2} & m_{3} \end{pmatrix} \) is the 3-j symbol, related to the Clebsch-Gordan coefficient by:

\[
\begin{pmatrix} j_{1} & j_{2} & j_{3} \hline m_{1} & m_{2} & m_{3} \end{pmatrix} = (\pm 1)^{j_{1}-j_{2}-m_{3}} \times \begin{pmatrix} j_{1} m_{1} & j_{2} m_{2} & j_{3} m_{3} \hline 2j_{3} + 1 & -1/2 \end{pmatrix}. \tag{12}
\]

The non-zero matrix elements from Eq. (11) satisfy the following selection rules: \( |J - 2| \leq J' \leq J + 2 \) and \( k' = k, k \pm 2 \).

In order to evaluate the values of \( \beta_{1,2} \), we consider the following pulse shape: \( \varepsilon_{2}^{2}(t) = \varepsilon_{0}^{2} \exp \left\{ -\frac{t^{2}}{\sigma^{2}} \right\} \). Considering the maximal pulse intensity to be \( I = 3 \times 10^{13} \text{W/cm}^{2} \) and duration \( \sigma = 20 \text{fs} \), we evaluate \( \varepsilon_{0} = 1.5 \cdot 10^{10} \text{V/m} \) and obtain \( \beta_{1} = -0.174 \) and \( \beta_{2} = -0.065 \).

In order to calculate the alignment factor \( \langle \cos^{2} \theta \rangle_{T} \) at temperature \( T \), one has to perform thermal averaging:

\[
\langle \cos^{2} \theta \rangle_{T}(t) = \sum_{J, \tau, m} \frac{\exp\left\{ -E_{J'}/k_{B}T \right\}}{Q_{\text{rot}}} \langle \cos^{2} \theta \rangle_{J\tau m}(t) . \tag{13}
\]

Here \( Q_{\text{rot}} \) is the rotational partition function, \( k_{B} \) is the Boltzmann’s constant, \( \langle \cos^{2} \theta \rangle_{J\tau m} \) denotes quantum thermal average over the molecular ensemble, and the indexes \( J, \tau, m \) denote the initial rotational state \( |J, \tau, m\rangle \) of a molecule in the ensemble. The quantum average is calculated using:

\[
\langle \cos^{2} \theta \rangle_{J\tau m}(t) = \langle \Psi(t)|\cos^{2} \theta|\Psi(t)\rangle, \tag{14}
\]

where \( \Psi(t) \) is found by solving Eqs. (8), (9), and (10) with the initial state \( \Psi(t = 0^{-}) = |J, \tau, m\rangle \). The right hand side in Eq. (14) can be easily calculated using the relation:

\[
\cos^{2} \theta = \frac{2D_{02}^{2}(\hat{R}) + 1}{3}. \tag{15}
\]

It is natural to consider the alignment of the para and ortho water molecules separately, since the laser pulse does not mix the para and ortho rotational states, i.e. if one considers an initial para rotational state and apply the pulse, only para rotational states are excited. The same holds for ortho rotational states. This is because the interaction term, \( H_{\text{ind}} \), in Eq. (4) includes:

\[
D_{02}^{2}(\hat{R}) \propto \langle \hat{R}|J = 2, k = 0, m = 0\rangle \times \langle \hat{R}|J = 0, k = 2, m = 0\rangle, \tag{16}
\]

which (according to Eq. (18) in the Appendix) have \( A_{1} \) symmetry. In order to have a transition from a para to ortho state, the following matrix element should not necessarily vanish: \( \langle \text{Para}|H_{\text{ind}}|\text{Ortho} \rangle \). Since the interaction term has a symmetry \( \Gamma_{\text{ind}} = A_{1} \), the para state has \( \Gamma_{\text{para}} = A_{1} \) or \( A_{2} \) symmetries, and the ortho has \( \Gamma_{\text{ortho}} = B_{1} \) or \( B_{2} \), direct product of the symmetries \( \Gamma_{\text{para}} \times \Gamma_{\text{ind}} \times \Gamma_{\text{ortho}} \) never contains \( A_{1} \) and therefore necessarily \( \langle \text{Para}|V_{\text{ind}}|\text{Ortho} \rangle = 0 \) (forbidden transition). Thus, the transitions between para and ortho states are forbidden and the sum in Eq. (13) includes either initial para states or initial ortho states separately.

IV. RESULTS

Applying a linearly polarized \( 3 \cdot 10^{13} \text{W/cm}^{2}, \) \( 20 \text{fs} \) laser pulse, we expect the molecular highest polarizability axis (i.e. the \( a \) axis in Fig. 1) to be aligned along the laser field polarization shortly after the pulse. Since the pulse is short compared to the typical rotational periods of the water molecule, it is considered as a delta-pulse. The calculated alignment factor \( \langle \cos^{2} \theta \rangle_{T} \) (Eq. (13)) is plotted in Fig. 3 as a function of time at temperature of 20K.
FIG. 3: The alignment factor after the laser pulse at temperature of 20K. Different dynamics of the ortho and para isomers is revealed during free propagation of the rotational wavepackets.

It is seen that the ortho and para rotational wavepackets evolve differently after the application of the pulse, and transient moments of simultaneous alignment and anti-alignment for the two species can be found. For instance, at $t = 2$ ps, the para molecules are aligned while the ortho molecules become anti-aligned. Naturally, the rotational energies of both ortho and para molecules are increased as the result of the pulse. Further selective control of the isomers can be achieved by applying an additional pulse (of the same intensity and duration, for simplicity) at $t = 1.9$ ps, when the ortho and para molecules are on the way of becoming anti-aligned and aligned, respectively. At this time, the alignment factors of both isomers are close to each other, and are a bit larger than $1/3$. The dynamics of the alignment factors after the second pulse is shown in Fig. 4 (again, for temperature of 20K).

It depicts a selective alignment (and anti-alignment) of the para molecules, while leaving the ortho modifications more or less isotropically distributed. Moreover, the total rotational energy of the ortho and para molecules is affected in a different way by the second pulse. While the rotational energy of the para molecules is further increased, the energy of the ortho molecules is reduced as the result of the action of the second pulse.

V. CONCLUSIONS

We considered interaction of the water molecules with strong and short off-resonant laser pulses and analyzed dynamics of rotational wavepackets of ortho and para spin isomers of water. We showed that these two forms of the water molecules experience distinct series of transient alignment and anti-alignment events, which leads to the angular separation of an initially isotropic mixture of nuclear spin modifications. Moreover, we showed that by applying a second, properly delayed laser pulse it is possible to suppress the rotational energy of one isomeric species, and leave it in a more or less isotropic state. On contrast, the other species will experience an enhanced series of transient alignments and anti-alignments. Such a selective alignment establishes grounds for the actual separation of ortho and para water isomers via interaction with additional spatially inhomogeneous static or time-dependent fields. This is a subject of our ongoing research.

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VI. APPENDIX- ROTATIONAL WAVEFUNCTIONS CLASSIFICATION

The eigenstates of the water molecule are classified using the $C_{2v}(M)$ Molecular Symmetry (MS) group [1]. Considering the water molecule eigenstate (Eq. (1)), we define the nuclei rotational and vibrational, as well as electronic degrees of freedom as a rovibronic eigenstate. We denote the rovibronic eigenstate symmetry as $\Gamma_{rne}$ (within the MS group), and the nuclear spin state symmetry as $\Gamma_{ns}$ (within the MS group). The water nuclear spin states are divided into either $S = 0$ or 1 nuclear spin states, denoted as para or ortho, respectively. The Pauli principle imposes symmetry limitations on the valid basis functions for expressing the internal molecular wavefunction, $\Phi_{int}$ (the entire molecular wavefunction, without the translational degrees of freedom). A rovi-
bronic state having $\Gamma_{\text{rov}}$ symmetry can be only combined with a nuclear spin state having $\Gamma_{\text{ns}}$ symmetry if the product of the two symmetries is an allowed symmetry for $\Phi_{\text{int}}$. In this Appendix we review the symmetries of the rotational eigenstates, and identify each rotational eigenstate as para or ortho.

We start by examination of the symmetry of the internal wavefunction, $\Phi_{\text{int}}$. The character table of the MS group is given in Table I. The group operations are: $E$, $(12)$, $(12)^*$, where $E$ is the identity operator, $(12)$ is the permutation of the Hydrogen nuclei operator, $E^*$ is the inversion operator (both for the nuclei and electron coordinates) and $(12)^*$ is the combination $(12)E^*$. In Table I only the symmetries $B_2$ (positive parity with respect to inversion) and $B_1$ (negative parity with respect to inversion) change sign under the operation $(12)$ (i.e. the permutation of the two Hydrogen nuclei, which are fermions). Therefore, they are the only allowed symmetries of the internal wavefunction.

Next, we classify the nuclear spin wavefunction. There are two possible spin functions for $s = 1/2$ proton spin: $\alpha = (1/2, 1/2)$ and $\beta = (1/2, -1/2)$, where $\alpha$ and $\beta$ correspond to spin up and spin down states, respectively. There are four possible spin configurations:

$$m = 1 : \alpha \alpha = \Phi_{\text{ns}}^{(1)}$$
$$m = 0 : \alpha \beta = \Phi_{\text{ns}}^{(2)} ; \beta \alpha = \Phi_{\text{ns}}^{(3)}$$
$$m = -1 : \beta \beta = \Phi_{\text{ns}}^{(4)} .$$

One should bear in mind that the spin functions are invariant to $E^*$, and also for them $(12)^* = (12)$. $\Phi_{\text{ns}}^{(1)}$ and $\Phi_{\text{ns}}^{(4)}$ are invariant to the group operations and have symmetry $A_1$ (see Table I). The characters of $\Phi_{\text{ns}}^{(2)}$ and $\Phi_{\text{ns}}^{(3)}$ (which transform together) are 2,0,2,0 for $E,(12), E^*, (12)^*$, respectively, i.e. $\Phi_{\text{ns}}^{(2)}$ and $\Phi_{\text{ns}}^{(3)}$ generate the representation $A_1 \otimes B_2$. The four nuclear spin wavefunctions therefore generate the representation $\Gamma_{\text{ns}} = 3A_1 \oplus B_2$.

| $C_{2v}(M)$ | $E$ | $(12)$ | $E^*$ | $(12)^*$ |
|------------|-----|-------|------|--------|
| Rotation $R^0$, $R_6^*$ | 1 | 1 | 1 | 1 |
| $A_2$ | 1 | 1 | -1 | -1 |
| $B_1$ | 1 | -1 | -1 | 1 |
| $B_2$ | 1 | -1 | 1 | -1 |

TABLE I: The character table of the $C_{2v}(M)$ MS group. $E$ is the identity operator, $(12)$ is the permutation of the Hydrogen nuclei operator, $E^*$ is the inversion operator (both for the nuclei and electron coordinates) and $(12)^*$ is the combination $(12)E^*$. The effect of the MS group on the rotational eigenstates is equivalent to the effect of the operations of the molecular rotation group $D_2$ (i.e. $R_0^0, R_6^*, R_8^*, R_6^*$) on the rotational eigenstates.

As the next step, we classify each rotational eigenstate. It turns out that the effect of the MS group on the rotational eigenstates is equivalent to the effect of the operations of the molecular rotation group $D_2$ (i.e. $R_0^0, R_6^*, R_8^*, R_6^*$) on the rotational eigenstates.

From Table I one can relate the $A_1$ and $A_2$ rovibrational wavefunctions with $S = 0$ nuclei spin, i.e. para water (the antisymmetric $S = 0$ wavefunction has a $B_2$ symmetry). Similarly, the $B_1$ and $B_2$ rovibrational wavefunctions are related with the $S = 1$ nuclei spin, i.e. ortho water (the symmetric $S = 1$ wavefunctions generate the representation $3A_1$). The statistical weight is therefore clear from the Table: there are three times more ortho species than para species (75% of the water is ortho and 25% is para).

Finally, Table III identifies the eigenstates of water molecule as para or ortho for various values of $J$ and $\tau$.
| Para(P)/Ortho(O) Symmetry J | τ |
|-----------------------------|---|
| P  | A₁ | 0 | 0 |
| O  | B₁ | 1 | −1 |
| P  | A₂ | 1 | 0 |
| O  | B₂ | 1 | 1 |
| P  | A₁ | 2 | −2 |
| O  | B₂ | 2 | −1 |
| P  | A₂ | 2 | 0 |
| O  | B₁ | 2 | 1 |
| P  | A₁ | 2 | 2 |
| O  | B₁ | 3 | −3 |
| P  | A₂ | 3 | −2 |
| O  | B₂ | 3 | −1 |
| P  | A₁ | 3 | 0 |
| O  | B₁ | 3 | 1 |
| P  | A₂ | 3 | 2 |
| O  | B₂ | 3 | 3 |

TABLE III: Para and ortho wavefunctions (here presented only till \( J = 3 \)) identification. The water asymmetric rotor eigenstates are \( |J, \tau, m\rangle \), where \( J \) is the angular momentum quantum number \( (J = 0, 1, 2, \ldots) \), and \( m \) is the quantum number of the angular momentum projected on the laboratory \( Z \) axis (since there exists no preferred direction in space, we choose here \( m = 0 \) without loss of generality). Index \( \tau \) numerates various \( 2J + 1 \) eigenstates arranged from lowest energy \( (\tau = −J) \) to highest energy \( (\tau = +J) \). The Table displays also the symmetry of the rotational eigenstates.

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