Complete alignment and orientation of atoms and molecules by stimulated Raman scattering with temporally shifted lasers

Y. B. Band and P. S. Julienne

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LETTERS TO THE EDITOR

COMMUNICATIONS

Complete alignment and orientation of atoms and molecules by stimulated Raman scattering with temporally shifted lasers

Y. B. Band

Department of Chemistry, Ben-Gurion University, Beer Sheva, Israel 84105 and Institute for Theoretical Atomic and Molecular Physics, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts 02138

P. S. Julienne

Molecular Spectroscopy Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899

Recent experiments have demonstrated that complete population transfer can be achieved in atoms and molecules using stimulated Raman scattering with time dependent fundamental and Stokes laser fields that are temporally delayed but partially overlapping.[1,2] The transfer efficiency in these experiments was unity when the process satisfied adiabatic passage criteria.[1-5] The high transfer probability was surprising because (a) the decay time of the excited intermediate state was very short compared to the laser pulse durations (and the fundamental and Stokes frequencies were tuned to resonance, so one might expect the excited intermediate state of the Raman transition to be populated), and (b) the atomic or molecular states contained internal angular momentum J, and therefore the Rabi frequencies for the fundamental and Stokes transitions depend on the magnetic quantum numbers M of the molecular states participating in the transitions for polarized laser fields; nevertheless, all the M states were transferred. Full density matrix calculations including the magnetic quantum number dependence of the Rabi frequencies[3] have been performed and adiabatic following descriptions[1,3,4] have been advanced to interpret and explain the unexpected experimental results. Alignment and orientation of the initial and terminal levels of the Raman transition as a function of pulse time displacement have been predicted.[3] However, the alignment and orientation was not complete using the scheme presented in Ref. 3.

It is the purpose of this Communication to propose some simple schemes for obtaining complete alignment and orientation of atoms and molecules using stimulated Raman scattering with temporally shifted laser fields. We shall work out the schemes for molecules; the atomic case is essentially identical. The idea is to totally move the population of a certain subset of the magnetic sublevels of an initial molecular state $\{ |v\Lambda JM\rangle, M = -J,...,J \}$ that is optically coupled to the excited intermediate and terminal states by the stimulated Raman scattering, with complete adiabatic passage of the subset of M states insured, while the remaining magnetic sublevels of the initial state are not optically coupled and therefore remain in the initial state.[6]

Here v is the set of internal state quantum numbers (e.g., the vibrational quantum number for a diatomic molecule), Λ is the projection of the angular momentum on the axis of the molecule, J is the angular momentum, and M is the projection of the angular momentum on the space fixed z axis (the polarization direction of the electromagnetic field for linearly polarized light). The critical point here is that all the states in the subset are totally transferred while the remain magnetic sublevels are unaffected. The simplicity and efficiency of the present method is striking. Alternative methods of creating aligned and oriented states using stimulated Raman scattering population transfer with fundamental and Stokes laser fields having different polarizations in the presence of magnetic fields have recently been proposed,[7] but the present scheme seems simplest.

The Liouville equation for the density matrix of the three level system (initial, excited intermediate, and terminal states labeled by g, a, and b, respectively) interacting with two optical time dependent pulses, including decay due to spontaneous emission, is given by[3]

$$\frac{d}{dt}\rho = -i[H(t),\rho] - \Gamma\rho, \tag{1}$$

where $H = H_0 - E(t)\cdot\mu$, H_0 is the molecular Hamiltonian, the electromagnetic field $E(t)$ is

$$E(t) = A(t)\exp[-i\omega_1 t] + B(t)\exp[-i\omega_2 t] + \text{c.c.}, \tag{2}$$

μ is the transition dipole moment matrix for the system,

$$\mu = \begin{pmatrix} \mu_{gg} & \mu_{ag} & \mu_{gb} \\ \mu_{ag} & \mu_{aa} & \mu_{ab} \\ \mu_{bg} & \mu_{ab} & \mu_{bb} \end{pmatrix} = \begin{pmatrix} 0 & \mu_{ag} & 0 \\ \mu_{ag} & 0 & \mu_{ab} \\ 0 & \mu_{ab} & 0 \end{pmatrix}, \tag{3}$$

and the decay matrix Γ has only the following nonvanishing elements: $\Gamma_{gg,aa}$, $\Gamma_{bb,aa}$, $\Gamma_{ag,ag}$, and $\Gamma_{ba,ba}$. The Rabi frequencies are given in terms of the transition dipole moments and field strengths by $\Omega_p(t) = A(t)\mu_{ag}/\hbar$ and $\Omega_s(t) = B(t)\mu_{ab}/\hbar$.

In actuality, the states g, a, and b have angular momentum denoted by J, J', and J'', respectively, so a mani-

Alignment with Linear Polarization
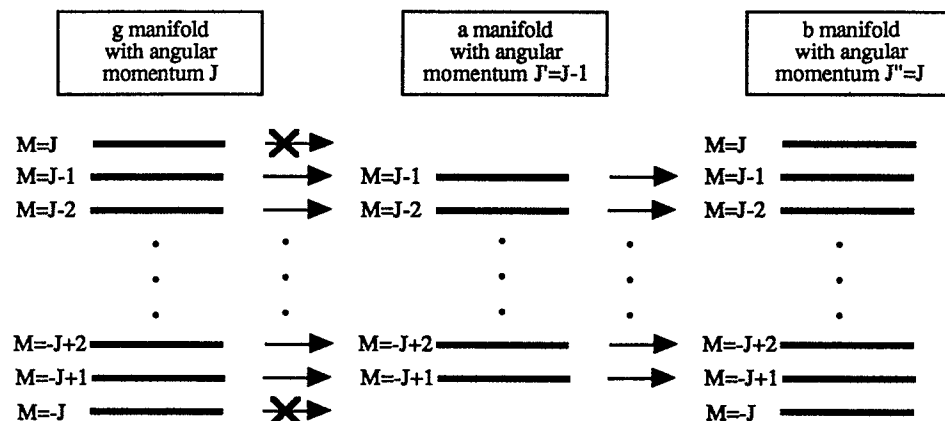

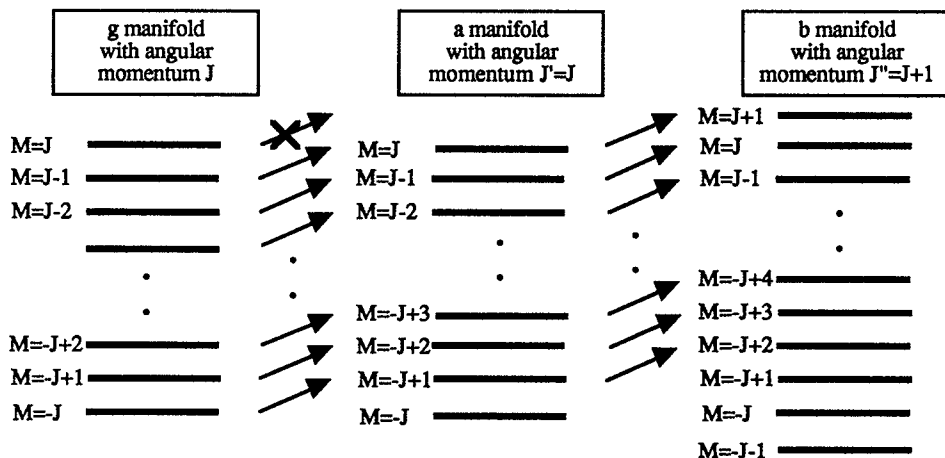## Orientation with Circular Polarization

FIG. 1. Alignment and orientation schemes using stimulated Raman scattering with temporally shifted laser beams giving rise to complete population transfer via adiabatic passage of the allowed transitions.

fold of magnetic quantum numbers exists for each state. Hence, the Rabi frequency for a particular molecular transition $v\Lambda JM\leftrightarrow v'\Lambda'J'M'$ for light with polarization q ($q = 0$, or ± 1), is given by[3]

$$\Omega_{1;v'J'M',vJM}(t) = A(t)\cdot\langle v'\Lambda'J'M'|\mu|v\Lambda JM\rangle/\hbar$$

$$= [0.5](8\pi\hbar\omega I_1/c)^{1/2}$$

$$\times\langle v'|d|v\rangle f(J'M'JM;\Lambda',\Lambda,q). \qquad (4)$$

Here the factor [0.5] is included because our definition of Rabi frequency involves A (not E which equals $2A$), I is the laser intensity in units of (photons/cm^2 s), d is the electronic transition dipole moment, $d = \langle\Lambda'|\mu|\Lambda\rangle$, and $f(J'M'JM;\Lambda',\Lambda,q)$ is the rotational line strength factor. This is similar for the Rabi frequencies $\Omega_{2;v''J''M'',v'J'M'}(t)$ involving the field at frequency ω_2. The Wigner–Eckart theorem can be invoked to write the rotational line strength factor as

$$f(J'M'JM;\Lambda',\Lambda,q) = (J1J'|M,q,M')S(J,J';\Lambda,\Lambda'). \qquad (5)$$

The reduced line strength factor S (or Hönl–London amplitude) is independent of M, M', and q and provides the selection rule for the molecular state transitions.[8] We assume here the same z axis defines the laser polarization q for both fundamental and Stokes fields and the quantization axes of all three molecular states.

If we assume parallel linear polarizations for the two lasers ($q = 0$), the line strength factors for P, Q, and R transitions ($J' = J - 1$, J, and $J + 1$, respectively) are proportional to

$$f_P\sim[J^2 - M^2]^{1/2}, \qquad (6)$$

$$f_Q\sim M, \qquad (7)$$

$$f_R\sim[(J + 1)^2 - M^2]^{1/2}. \qquad (8)$$

It is evident from Eq. (6) that quite generally, for arbitrary J and $\Lambda\leftrightarrow\Lambda'$ transitions, the transition from the $|M_J| = J$ level vanishes for P transitions, i.e., $J^2 - M^2 = 0$ in Eq. (6). Therefore, for transitions from g, a, and b levels with angular momentum J, $J' = J - 1$, and $J'' = J$, respectively, using linearly polarized light, all the population in the

$|M_J| = J$ levels of g are left in the initial state, whereas all other M_J levels will be transferred to the terminal level if the fundamental and Stokes fields are properly timed and sufficiently intense to insure adiabatic passage (even cw lasers intensities are sufficient if the frequencies are resonant or nearly so). Hence the resulting alignment parameter for state g is $A_2 = -2$, i.e., the state is fully aligned. This scheme is illustrated in the top part of Fig. 1. For the special case of $J = 1$, the only final state populated is $J'' = 1$, $M'' = 0$, i.e., the final state is aligned with $A_2 = 1$, with all the population in the ground state $M = 0$ level having been transferred. If two Q branch transitions ($J = J' = J''$) are used with two linearly polarized lasers, Eq. (7) shows that population in the $M = 0$ level of the ground state will not be moved. Thus, the g state will be left with $A_2 = 1$ alignment, regardless of J, whereas the terminal state will have population missing for $M = 0$. It is evident that several variations are possible by varying q and branch for the two transitions.

The rotational line strength factor for right circularly polarized light ($q = +1$) for Q branch transitions is proportional to

$$f_Q \sim [(J - M)(J + M + 1)]^{1/2}. \tag{9}$$

Hence, if the Q branch transition is allowed, transitions involving all M_J except $M_J = J$ will occur, but the Rabi frequency for $M_J = J$ vanishes. If $J'' = J + 1 = J' + 1$, the transfer of the population from all M_J except $M_J = J$ can be complete, leaving the state g fully oriented. This scheme is pictorialized in the bottom of Fig. 1.

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