

Selecting molecules in the vibrational and rotational ground state by deflection

P. Domokos^a, T. Kiss^b, and J. Janszky

Research Institute for Solid State Physics and Optics, 1525 Budapest, P.O. Box 49, Hungary

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Abstract. A beam of diatomic molecules scattered off a standing wave laser mode splits according to the rovibrational quantum state of the molecules. Our numerical calculation shows that single state resolution can be achieved by properly tuned, monochromatic light. The proposed scheme allows for selecting non-vibrating and non-rotating molecules from a thermal beam, implementing a laser Maxwell's demon to prepare a rovibrationally cold molecular ensemble.

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Manipulating the motion of massive particles by using the mechanical effect of light has been a rapidly developing field since the discovery of lasers. Understanding the fine details of the light–matter interaction on a microscopic level contributed much to the advent of impressive applications, including laser cooling and trapping of atoms and atom optics [1]. The adaptation of these techniques for molecules would undoubtedly open new perspectives. Recently, pioneering experiments have been reported in this direction, for trapping [2,3] and decelerating [4] molecules or for interferometry [5], and also for fundamental tests of quantum mechanics [6] with molecules. It is, however, non-trivial to extend the optical methods to molecules because of the complexity of their energy level structure. Optical cooling of molecules [7] would require new approaches, for example, such as coupling the particle to a far-detuned light field inside an optical cavity [8].

The essential difference stems from the additional mechanical degrees of freedom. The vibrational and rotational modes lead to densely spaced levels. Typical excitations are orders of magnitude smaller than the electronic transitions. A usual thermal beam consists then of molecules in various rovibrational quantum states. Cooling translation, vibration, and rotation simultaneously is a difficult problem; a complex scheme with multiple laser beams has been proposed in [9]. However, there is another option to obtain molecules with lower energy: to sort the thermal ensemble by means of state-sensitive interactions. A Stern-Gerlach-type experiment with inhomogeneous

microwave field has been first performed to separate the rotational states constituting the resonant transition within a given vibrational band [10]. Rotational state dispersion has been shown to be present in various molecular optics settings using far-detuned laser fields [11]. In this paper we show that a Maxwell demon like operation can be achieved in a different regime of the molecule-light interaction. That is, a single rovibrational state, possibly the ground state, can be filtered from a thermal beam. Owing to the high degree of control facilities, the proposed scheme can be applied to a quite general class of molecular states.

Deflection of a CS₂ and I₂ beam using the nonresonant dipole force has recently been demonstrated [12]. The laser was well detuned from a given electronic transition and all the molecules experienced very nearly the same nonresonant dipole force regardless of what their internal quantum state was. In order to accommodate the nonresonance condition for all the transitions, detuning on the scale of the electronic transition frequency was applied. More sophisticated control of the mechanical effect of light could be obtained with a laser whose bandwidth is comparable with the linewidth of the rovibrational transitions in the spectrum. With detunings on the order of the rotational level-spacing, less laser power is needed to have the same mechanical effect.

The reaction of atoms and molecules to radiation is usually described by the frequency-dependent polarizability tensor. This is a good approximation for a transparent medium, when the radiation frequency is far away from any resonances in the matter. The polarizability can then be calculated from the quantum mechanical mean value of the dipole operator, usually by employing perturbation theory. This approach is sufficient to interpret the

^a *Present address:* Institute für Theoretische Physik, Universität Innsbruck, Technikerstrasse 25, 6020 Innsbruck, Austria. e-mail: peter.domokos@uibk.ac.at

^b *Also at:* University of Pécs, Ifjúság u. 6, 7624 Pécs, Hungary.

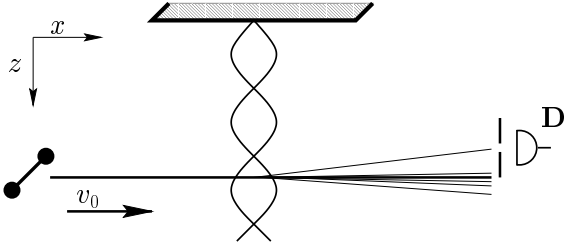


Fig. 1. Scheme of deflecting molecules quantum state selectively. A beam of diatomic molecules with velocity v_0 is scattered off a monochromatic, standing wave laser mode. The detector **D** measures the deflected beam intensity along the z -axis.

deflection experiment [12], and many other schemes for manipulating molecular motion [13,14]. However, the simple concept of a state-independent polarizability does not hold when the monochromatic laser frequency is close to some individual rovibronic resonances. In this case one must return to a more fundamental quantum mechanical treatment relying on the dipole operator itself. Otherwise the standard theoretical framework for treating molecular optics [15], that is the adiabatic separation of the rovibrational modes and the center-of-mass motion can be used.

We consider a beam of diatomic molecules (see Fig. 1) moving with velocity v_0 in the x -direction and crossing at a right angle a CW standing-wave laser mode. The interaction energy, in the dipole approximation, reads $V = -\hat{\mathbf{d}} \cdot \mathbf{E}$. The electric field is linearly polarized, and its spatial variation is given by $\mathbf{E} = \mathbf{E}_0 \exp[-(x^2 + y^2)/2w^2] \cos kz \cos \omega t$. The molecules experience a force proportional to the gradient of the inhomogeneous interaction energy. Due to this light force, the molecules accumulate transverse momentum, Mv_z . However, the displacement in this direction is negligible during the interaction (Raman–Nath regime). The laser-induced potential is assumed to be much smaller than the kinetic energy; hence the longitudinal velocity $v_x \equiv v_0$ remains constant during the whole passage.

The spectrum of the diatomic molecule in the Hund’s case a is defined [16] by the Hamiltonian

$$H_0 = \sum E(n, \nu, J, M, \Omega) |n\nu JM \Lambda \Sigma\rangle \langle n\nu JM \Lambda \Sigma|, \quad (1)$$

where n labels the electronic states, ν is the vibrational, J and M are the usual rotational quantum numbers, Λ and Σ are the projections of the total electronic angular momentum and spin on the intermolecular axis, respectively, and Ω is their sum. Nuclear spin is ignored; however, we will comment on this point later on. Within the Born–Oppenheimer approximation, the electronic and nuclear wave functions can be separated as follows

$$|n\nu JM \Lambda \Sigma\rangle = \Phi_{n\Lambda\Sigma}(\tau_e, r) R_{n\nu}(r) u_{JM\Omega}(\theta, \phi), \quad (2)$$

where the variable τ_e incorporates all the electron coordinates, r is the internuclear distance, and θ and ϕ define the orientation of the molecule. The electronic spin part of the quantum state is omitted. The energies for low ν

and J are usually approximately given by the expression

$$E(n, \nu, J, M, \Omega) = E_{\text{el}}(n) + \hbar\omega_e(\nu + 1/2) - \hbar\omega_e x_e(\nu + 1/2)^2 + B_\nu[J(J+1) - \Omega^2] - D_\nu[J(J+1) - \Omega^2]^2, \quad (3)$$

where the first term is the electronic energy, the next two terms describe the anharmonic vibration, and the last two correspond to the energy of a symmetric top (the standard notation [16,17] is used).

Let us suppose that only the fundamental “f” and one excited “e” electronic states are effectively coupled by the electromagnetic mode. The interaction Hamiltonian can be restricted to

$$V = -\mathbf{E}_0 e^{-(x^2+y^2)/2w^2} \cos kz \cos \omega t \times \sum_{\nu JM} \sum_{\nu' J' M'} \mathbf{d}_{f\nu JM}^{e\nu' J' M'} |f\nu JM\rangle \langle e\nu' J' M'| + \text{h.c.} \quad (4)$$

The electric field \mathbf{E}_0 is linearly polarized in an arbitrary direction perpendicular to the mode axis z . It is enough to consider the dipole moment in the direction of the polarization. We resort to the Franck–Condon approximation to calculate the matrix elements of the dipole operator, *i.e.*, the induced electric dipole moment is supposed to be independent of the internuclear distance. This separation is not fundamental, but only convenient for purposes of discussion. The matrix element can then be decomposed into three factors, $d_{f\nu JM}^{e\nu' J' M'} = d_f^e R_\nu^{J' M' \Omega'} L_{JM\Omega}^{J' M' \Omega'}$. The electric dipole is given by the integral

$$d_f^e = \int \Phi_e^*(\tau_e, r) \hat{\mathbf{d}} \Phi_f(\tau_e, r) d\tau_e, \quad (5)$$

averaged over the nuclear distances r . The next term,

$$R_\nu^{J' M' \Omega'} = \int R_{e\nu'}^* R_{f\nu} r^2 dr, \quad (6)$$

is the vibrational overlap factor (the square root of the Franck–Condon factor) that we will calculate using Morse-potential eigenfunctions. Finally, a rotation-dependent factor follows which can be analytically evaluated for the symmetric top eigenfunctions. Two cases can be distinguished:

$$L_{JM\Omega}^{J' M' \Omega'} = \int u_{J' M' \Omega'}^* u_{JM\Omega} \begin{Bmatrix} \cos \theta \\ \sin \theta \end{Bmatrix} \sin \theta d\theta d\phi, \quad (7)$$

where $\cos \theta$ is used for transitions with $\Delta\Omega = 0$, and $\sin \theta$ is used for transitions with $\Delta\Omega = \pm 1$.

The Hamiltonian, in the rotating wave approximation, can be transformed to the form

$$\hat{H}_{\text{eff}} = \sum_{\nu JM} \sum_{\nu' J' M'} \bar{E}_{\nu JM}^{J' M' \Omega'} \times (|f\nu JM\rangle \langle f\nu JM| + |e\nu' J' M'\rangle \langle e\nu' J' M'|) + \frac{1}{2} \hbar \delta_{\nu JM}^{J' M' \Omega'} (|e\nu' J' M'\rangle \langle e\nu' J' M'| - |f\nu JM\rangle \langle f\nu JM|) + \hbar g_{\nu JM}^{J' M' \Omega'} f(\mathbf{R}) (|f\nu JM\rangle \langle e\nu' J' M'| + \text{h.c.}). \quad (8)$$

The first term defines the energy zero level for each rovibronic transition $|f\nu JM\rangle \leftrightarrow |e\nu'J'M'\rangle$. The second term is the corresponding detuning from the laser frequency, $\hbar\delta_{\nu JM}^{\nu'J'M'} \equiv E(e\nu'J'M') - E(f\nu JM) - \hbar\omega$. The coupling strength is given by $g_{\nu JM}^{\nu'J'M'} \equiv \mathbf{d}_{f\nu JM}^{e\nu'J'M'} \mathbf{E}_0/\hbar$. The mode function $f(\mathbf{R})$ collects the spatially varying factors. The coupling constant can be expressed in terms of measurable quantities as

$$\left(g_{\nu JM}^{\nu'J'M'}\right)^2 = \frac{3(\lambda_{f\nu J}^{e\nu'J'})^3}{16\pi^2\hbar c} \Gamma_{f\nu J}^{e\nu'J'} \frac{(2J'+1)|L_{JM\Omega}^{J'M'\Omega'}|^2}{S(J,J')} \frac{I}{A}, \quad (9)$$

where $\Gamma_{f\nu J}^{e\nu'J'}$ is the natural linewidth, and $S(J,J')$ is the Hönl-London factor. The laser parameters are the intensity I in units of photons/second and the effective beam area A .

The fine tuning of the laser is the crucial point of our scheme. The laser induces nonresonant interactions if the condition $(\delta_{\nu JM}^{\nu'J'M'})^2 \gg (g_{\nu JM}^{\nu'J'M'})^2$ holds for each transition. The laser is detuned from all the transitions, $\delta_{\nu JM}^{\nu'J'M'} > \Gamma_{\nu JM}^{\nu'J'M'}$, and the smallest detunings are on the order of the rotational level spacing B_ν . The nonresonance condition on this scale limits the coupling $g_{\nu JM}^{\nu'J'M'} \ll B_\nu$, and thus the laser intensity.

The molecules are initially in the electronic ground state, and the vibrational and rotational states are populated according to a thermal distribution. The Hamiltonian (8) describes the laser-induced dynamics of the molecule except for the translational motion. Interaction with the vacuum modes yielding spontaneous emission was omitted, since the upper electronic states are not populated by the nonresonant excitation. The translational motion is treated classically in our model. The center-of-mass (CM) coordinate appears as a parameter in \hat{H}_{eff} via the mode function $f(\mathbf{R})$. Motion in the y -direction is irrelevant with respect to the deflection scheme. We insert $v_0 t$ in the coordinate x , thus making the problem explicitly time-dependent. For slow enough motion, the system evolves adiabatically. In the nonresonant limit the eigenstates of \hat{H}_{eff} , which can be viewed as dressed rovibronic states, are close to those of the unperturbed \hat{H}_0 . Transitions can be neglected between these eigenstates. The eigenenergies are shifted (AC Stark effect) by an amount that can be well approximated by taking only the most significant transition into account for every rovibronic state. In fact, this approximation is equivalent to the decomposition of H_{eff} into 2 by 2 blocks. Corrections can be generated systematically by extending the block size with the inclusion of weaker and weaker couplings. The consistency of this method is based on the hierarchy among the couplings that can be set up according to the $g_{\nu JM}^{\nu'J'M'}/\delta_{\nu JM}^{\nu'J'M'}$ ratios. This is not a perturbational expansion: whenever an additional coupling term in \hat{H}_{eff} is considered, its contribution is completely included. In a first approximation,

the energy associated with the state $|f\nu JM\rangle$ reads

$$E_{\text{dressed}}(f\nu JM) = \bar{E}_{\nu JM}^{\nu'J'M'} \pm \hbar\sqrt{(g_{\nu JM}^{\nu'J'M'})^2 f(\mathbf{R})^2 + (\delta_{\nu JM}^{\nu'J'M'})^2/4}, \quad (10)$$

where $\bar{E}_{\nu JM}^{\nu'J'M'}$, $g_{\nu JM}^{\nu'J'M'}$, and $\delta_{\nu JM}^{\nu'J'M'}$ are the parameters of the transition to the strongest coupled upper rovibronic state $|e\nu'J'M'\rangle$. The sign is determined by the sign of the detuning. During the passage the molecules acquire transverse velocity due to the nonresonant dipole force, given by the gradient of the energy equation (10). We predict the occurrence of different trajectories labeled by the quantum numbers of the relevant rovibrational states. The quantum state dependent deflection angles read

$$\alpha(\nu JM) = \frac{v_z}{v_x} \approx \frac{v_{\text{rec}} (g_{\nu JM}^{\nu'J'M'})^2 l}{v_x^2 \delta_{\nu JM}^{\nu'J'M'}}, \quad (11)$$

where $l = \sqrt{A}$ is the effective interaction length, $v_{\text{rec}} = \hbar k/M$ is the photon recoil velocity.

In the following, we analyze the complex deflection pattern on the example of the Na₂. The ground electronic state $X^1\Sigma_g^+$ is coupled to the state $A^1\Sigma_u^+$ by a dipole-allowed optical transition with $E_{\text{el}} = 17541 \text{ cm}^{-1}$ [18]. Both states are singlet; hence the electron spin plays no role ($\Sigma = \Sigma' = 0$). Since $\Lambda = \Lambda' = 0$, the selection rules $J' = J \pm 1$, $M' = M$ apply (the rotational Q branch is missing) and the Hönl-London factor is $S(J,J') = J + 1$. According to theoretical calculations [19], confirmed by experiments [20], the electric dipole is $|\mathbf{d}_f^e|^2 = 14$ in atomic units.

Suppose the laser is tuned close to resonance with the rovibronic transition $f, \nu = 0, J = 0, M = 0 \leftrightarrow e, \nu' = 6, J' = 1, M' = 0$ (with energy $E_{\text{el}} + 666.136 \text{ cm}^{-1}$, and natural linewidth $\Gamma = 3.4 \times 10^{-5} \text{ cm}^{-1}$). The $0 \leftrightarrow 6$ vibrational transition is associated with one of the largest Franck-Condon factors ($|R_0^6|^2 = 0.113$). The neighboring transitions are represented in Figure 2a. For a temperature of 1000 K, the distribution can be truncated at 10 vibrational and 100 rotational levels. As the coupling strength is much less than B_ν , resonances that are as far as the rotational line separation ($B_\nu = 0.155 \text{ cm}^{-1}$) can be safely neglected. Thus a molecule has a chance to be deflected if its rovibrational state is involved in the transitions shown in Figure 2a. The adjacent lines are separated by at least 0.05 cm^{-1} ; hence $g \approx 1.5 \times 10^{-3} \text{ cm}^{-1}$ can be chosen ($\Gamma \ll g \ll B_\nu$). Focusing the standing wave to $l \approx 50 \mu\text{m}$, the necessary field intensity is $I \approx 0.3 \text{ mW}$. The Raman-Nath regime implies a purely geometrical condition on the deflection angle α that is limited by $\alpha < \lambda/l \approx 10^{-2}$ radian. The velocity is set at $v_0 = 500 \text{ m/s}$, yielding an interaction time of 100 ns. The probability of a spontaneous emission event is less than 10%; hence neglecting the scattering force is justified. Deflection angles according to the above parameters are shown in Figure 2b as a function of the laser frequency. Each curve belongs to a single rotational-vibrational state. The resonant region around each transition is darkened. Outside these regions the

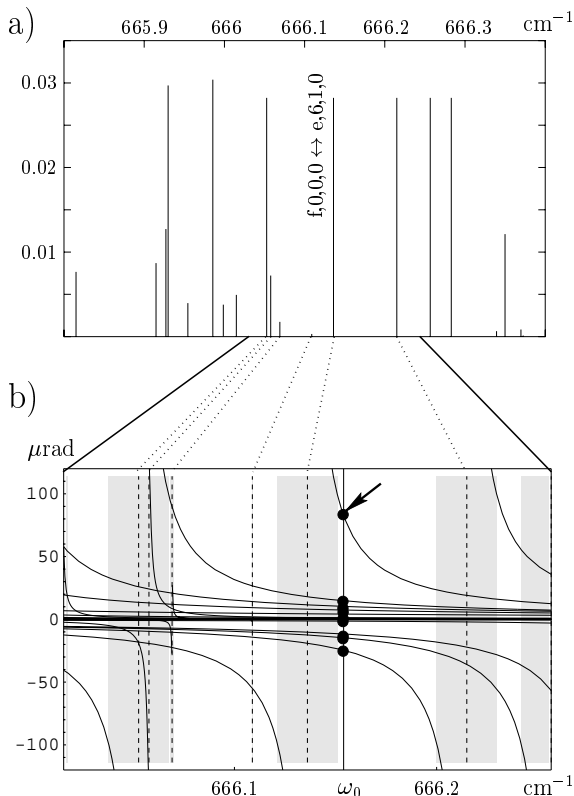


Fig. 2. (a) The spectrum of Na_2 around the transition $f, \nu=0, J=0, M=0 \leftrightarrow e, \nu'=6, J'=1, M'=0$. E_{cl} is subtracted from the frequencies. (b) Deflection angles as a function of the laser frequency. Dashed vertical lines correspond to the transitions. Outside the grey regions $\delta_{\nu,J,M}^{\nu',J',M'} > 10g_{\nu,J,M}^{\nu',J',M'}$ thus the non-resonance condition is fulfilled. At the specific laser tuning ω_0 several deflection angles occur (points). The arrow indicates the deflection of molecules in the vibrational-rotational ground state.

detuning is one order of magnitude larger than the corresponding coupling constant, thus the nonresonance condition is fulfilled. For the laser frequency ω_0 at the solid vertical line, the detuning will be $\delta \approx 0.02 \text{ cm}^{-1}$. The points represent the observable deflection angles. The arrow indicates the angle corresponding to the rotational-vibrational ground state (approximately $100 \mu\text{rad}$; for comparison, a single photon recoil would cause $35 \mu\text{rad}$). According to Figure 2b, several deflected partial beams are present simultaneously, which could be resolved depending on the beam divergence and on the detection technique. We note that a deflected beam does not correspond to a single quantum state, as we did not include in our model all degrees of freedom of a molecule, *e.g.* the nuclear spin was neglected. Thus the deflected beam contains all possible nuclear spins, altogether 6 states.

The center-of-mass motion has been described in terms of classical trajectories in our model, which offered an intuitive picture with focus on the more essential internal part of the dynamics. We avoided, however, some details that would degrade the predicted signal in an experimental realization. There are several mechanisms giving rise to a broadening of the deflected partial beams. First of all, we

assumed a well-defined longitudinal velocity v_x . It follows from (11) that the double the relative variance in v_x equals the relative variance of the deflection angle. Still we can safely tolerate a few percent uncertainty which can easily be achieved, even without velocity selection, by using supersonic beams. Next, the molecule has to cross the field in a region where the interaction strength, *i.e.* $f(\mathbf{R})$, is approximately uniform. The deflection angle being proportional to $\sin 2kz$, the crucial part in this respect is the direction z . The variation $2k\Delta z = \pm 0.45$ around $2kz = \pi/2$ amounts to 10% variation in the final deflection angle. However, the confinement of the beam to a size smaller than the wavelength causes diffraction. The transverse velocity that was supposed to be $v_z = 0$ broadens to a distribution with a width of $\hbar/(2M\Delta z) = v_{\text{rec}}/(2k\Delta z) \approx v_{\text{rec}}$. Deviations in the direction y from the center line $y = 0$ do not induce such drastic diffraction since the Gaussian beam waist is several orders of magnitude larger than the wavelength. Therefore $v_y \approx 0$ remains a good approximation. Finally, the spontaneous photon emission is accompanied by a recoil that introduces a randomness in the deflection angle. When the spontaneous emission probability is kept below 10%, the occurrence of several photon recoil kicks can practically be excluded. Similarly to the diffraction problem in the z -direction, the single photon recoil raises the question of resolution. That is, the deflection broadening, at most v_{rec}/v_x , must be dominated by the deflection angle given in (11). Note that these two quantities were $35 \mu\text{rad}$ and $100 \mu\text{rad}$ in our numerical example.

The effect of spontaneous emission is, however, a bit more complex than just broadening the velocity distribution. Following a spontaneous jump, the molecule exits the two states of the quasiresonant transition and the process of accumulating transverse momentum is interrupted. This gives rise to a weak background between the zero and maximum deflection angles. The suppression of this background to the possible minimum is the guiding principle when choosing the appropriate detunings. In fact, this imposes the ultimate condition on the generalization of the proposed scheme to other molecules. The basic ingredient of our scheme is the “isolated line” in the spectrum. That is the spectral density must leave enough space to tune the laser quasiresonant with a single transition without introducing photon scattering. Roughly, an order of magnitude between the detuning $\delta_{\nu,J,M}^{\nu',J',M'}$ and the linewidth $\Gamma_{\nu,J,M}^{\nu',J',M'}$, and also an order of magnitude between $\delta_{\nu,J,M}^{\nu',J',M'}$ and the distance to the adjacent transitions in the spectrum is necessary. In Na_2 , being a light diatomic molecule, we had three orders of magnitude difference between Γ and the relevant transition line separations. This suggests that the non-resonance condition can also be fulfilled for heavier diatomic or polyatomic molecules as long as the rotational constant B_ν is not less than one tenth of that of the sodium we considered here. Decisive conclusions on the applicability cannot be drawn without the detailed knowledge of the spectrum. Nevertheless, it may always be possible to find accidentally well isolated lines associated with the ground rovibrational state of the molecule.

In conclusion, we have shown that it is possible to deflect diatomic molecules quantum state selectively. The very high sensitivity originates from the monochromatic laser field that can be finely tuned within the rovibrational band. The laser-induced coupling of the vibrational and rotational modes to the center-of-mass translation results in quantum correlations between the different motional degrees of freedom. No analogue of this type of correlations exists in atom optics. There only the quantized light mode (quantum prism [21]) or the internal electronic state (optical Stern–Gerlach effect [22]) can be entangled to the translational motion. By contrast to atoms, however, in a usual beam of molecules various rovibrational states are thermally populated; hence the possibility of selecting the rovibrationally cold molecules has important practical consequences. This arrangement may be integrated in the preparation stage of experiments performing coherent manipulation with molecular beams. The process still cannot be called cooling, since thermal equilibrium is not maintained throughout. In fact, it is a laser Maxwell’s demon [23], which is not in contradiction to the second law of thermodynamics if the momentum exchange with the laser is correctly taken into account [24]. The Maxwell’s demon for molecules, beyond its unusual statistical properties, would be of great practical value for preparing a sample of non-vibrating and non-rotating molecules.

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