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Confining CO molecules in stable orbits

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Abstract

A scheme is presented for the confinement of neutral molecules in stable orbits on the basis of the linear Stark effect in a properly designed electrostatic trap. Based on Stark shift measurements in electric fields up to 140 kV/cm, an electrode configuration which is, under realistic experimental conditions, capable of confining metastable CO($a^3\Pi$) molecules with velocities up to 22 m/s is described. Metastable CO molecules can be laser-prepared inside the trap and their trajectories can be directly visualized via the spatially resolved detection of their fluorescence. © 1997 Elsevier Science B.V.

1. Introduction

Trapping atoms using laser cooling, together with techniques like magnetic trapping and evaporative cooling, has led to numerous exciting results over the last decade, with the realization of Bose–Einstein condensation as the most spectacular achievement [1]. Laser-cooling schemes are applicable to atoms since closed multi-level systems can be realized and many consecutive absorption–emission cycles can be induced. This leads to a significant momentum transfer from the photons of the laser to the atom, resulting in a change of the velocity of the atom. Trapping in one, two or three dimensions has been realized for many different atoms by now [2,3].

The trapping of neutral molecules has not been demonstrated yet. The above-mentioned laser-cooling schemes fail for molecules, even though multi-level systems that are electronically and rotationally closed can be found for molecules as well. The unavoidable off-resonant fluorescence decay of molecules from the excited state to other vibrational levels in the electronic ground state hampers efficient momentum transfer. As

a consequence, only the deflection of molecules over small angles using the sequential absorption of a limited number of photons is demonstrated [4]. Recently, a complex scheme to overcome this problem has been proposed for laser cooling Cs₂ (effectively a 21-level system) requiring an exceptional laser system [5].

Other schemes for trapping molecules have been investigated. Magnetic deflection has been realized for several molecules in the past [6,7], and mechanisms for magnetic [8] and electrostatic trapping [9] have been proposed. It has also been suggested that extremely high laser powers might be used to induce a dipole moment in polarizable molecules, leading to trapping of these molecules in the laser beam [10].

In this Letter we outline a scheme by which neutral metastable CO molecules can be confined in an appropriately shaped DC electric field on the basis of their linear (first-order) Stark effect. The scheme is based on the fact that the electric field between two coaxial electrodes scales with the distance r from the axis as $1/r$. This implies that molecules with a linear Stark effect experience a force that scales as $1/r^2$, and can therefore be captured in stable “planetary” orbits.

Metastable CO($a^3\Pi$) molecules are an ideal test candidate, as these can be laser-prepared inside the trap and as their trajectories can be directly monitored via the spatially resolved detection of their fluorescence.

In an inhomogeneous electric field, neutral molecules experience a force proportional to the gradient of the Stark energy. For molecules in electronic states with a large permanent electric dipole moment such inhomogeneous electric fields can therefore be used to efficiently deflect or focus these molecules. As the force acting on a molecule in a given quantum state depends on its rotational quantum numbers, inhomogeneous electric fields are suitable for state-selection. Often hexapole state-selectors are employed, although a variety of other electrode configurations has been proposed and experimented with [11]. In most cases the focusing performance is based on the first-order, i.e. low-field or linear, Stark effect. Recently, it has been demonstrated that polar molecules in extremely high inhomogeneous electric fields, molecules in so-called “pendular” states, can be state-selected as well [12]. In this Letter it was suggested that the electrode configuration under study, was also a promising candidate for the trapping of polar molecules. Other papers have elaborated somewhat further on this scheme using an electrode configuration similar to the one proposed here [13,14].

Rather than using “pendular” states, we propose confinement in stable orbits using the linear (first-order) Stark effect. A promising molecule for confinement using the linear Stark effect is the CO molecule. CO in its electronic ground state has only a small dipole moment, and, as it is in a $^1\Sigma^+$ state, does not experience a first-order Stark effect. CO in its first electronically excited state, the metastable $a^3\Pi$ state, has a relatively large dipole moment (1.37 D [15]) and does experience a linear Stark effect [15,16]. Metastable CO ($a^3\Pi$) molecules can therefore be efficiently deflected and focused using inhomogeneous electric fields [17]. CO molecules can be prepared in single quantum levels of this metastable state via laser excitation on the spin-forbidden $a^3\Pi(v=0) \leftarrow X^1\Sigma^+(v=0)$ transition of CO around 206 nm [16,17]. If the laser preparation of metastable CO is performed inside a DC inhomogeneous electric field, the force on the molecule is

suddenly switched on at the moment of excitation and acts on the molecule as long as it is in the metastable state. The lifetime of CO ($a^3\Pi$) is strongly quantum level dependent and ranges from 3–200 ms [18] for levels that can be readily laser prepared.

For the design of an electrostatic trap, it is important to know the exact dependence of the Stark shift on the electric field strength, as this will determine the trajectories of the trapped molecules. Although the Stark shift of the individual quantum levels in the $a^3\Pi$ state of CO can be calculated using known spectroscopic data, it is a priori not clear if the Stark shift will follow theory in the high electric fields that are necessary for confinement because neighbouring quantum levels of perturbing electronic states might interfere in these extremely high fields, thereby complicating the exact field behaviour.

2. Experimental

To verify the behaviour of the Stark shift as a function of electric field, CO $a^3\Pi(v=0) \leftarrow X^1\Sigma^+(v=0)$ excitation spectra are recorded as a function of the electric field in which the metastable CO molecules are prepared. The pulsed molecular beam machine used for these experiments has been described in detail elsewhere [17]. The radiation used to prepare CO in the metastable state is obtained via frequency-tripling the output of a pulsed dye amplified cw-ring dye laser system. This system produces pulses of 5 ns duration with a high spectral brightness (0.5 mJ/pulse in 150 MHz bandwidth), by which the spin-forbidden transition in CO can be efficiently induced. The laser excites the CO molecules in a homogeneous DC electric field that is applied between two parallel, gold coated electrodes placed 2.06(1) mm apart. This set-up allows high electric fields using relatively low (≤ 30 kV) voltages. The spectral resolution of the preparation laser system is sufficient to resolve the splittings of the various M -components in CO ($a^3\Pi_1, v=0, J=1$) even in electric fields as low as 1 kV/cm. Detection of the metastable CO molecules is performed using REMPI via the $b^3\Sigma^+ \leftarrow a^3\Pi$ transition in a field-free region, about 65 cm downstream from the excitation region.

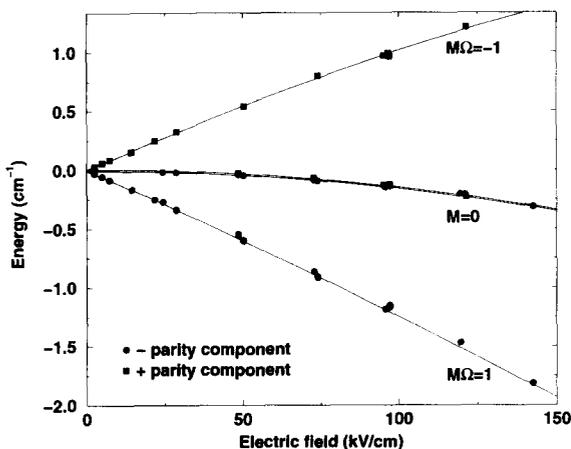


Fig. 1. Measured splitting of the different M -components of the CO $a^3\Pi_1$ ($\nu = 0$, $J = 1$) Λ -doublet as a function of the applied electric field. The vertical axis shows the position of the energy levels relative to the center of the Λ -doublet in zero electric field. The smooth curves indicate the calculated Stark shift.

3. Results and discussion

The behaviour of the different M -components of the CO ($a^3\Pi_1$, $\nu = 0$, $J = 1$) level in the electric field as deduced from the measured excitation spectra is shown in Fig. 1. Plotted is the energy of the various Stark components versus the applied electric field. In zero electric field the splitting between the Λ -doublet levels, the lower with $-$ parity and the upper with $+$ parity, is 394 MHz [15]. The origin of the vertical axis in Fig. 1 is taken at the center of the Λ -doublet in zero electric field. In the electric field each of the Λ -doublet levels is split in a component with $|M\Omega| = 1$ and a component with $M = 0$ (components originating from the $-/+$ parity level are indicated in the figure with circles/squares). The $M = 0$ component does not show a first-order Stark effect, and in low electric fields this level is therefore not significantly shifted. Fig. 1 shows that the components with $|M\Omega| = 1$ shift nearly linearly with the electric field up to field strengths of 50 kV/cm. In high electric fields it is observed that the $M = 0$ components shift over a few tenths of a cm^{-1} . This shift is caused by the second-order Stark effect, originating from the coupling of the $J = 1$ level with other rotational levels. Although the second-order Stark effect is largest for $M = 0$, it contributes significantly to the shift of the $|M\Omega| = 1$ components as well (see Fig. 1). The absolute value

of the Stark shift is largest for the $M\Omega = 1$ component (originating from the lower Λ -doublet level), due to the combined effect of the first- and second-order Stark effect. A total shift of 1.81 cm^{-1} is observed in an electric field of 140 kV/cm for this level. The CO molecules in this quantum level are therefore good candidates for confinement in stable orbits.

The smooth curves indicated in Fig. 1 show the calculated Stark shift of the different components, obtained using the known value of the dipole moment and the rotational energy level structure [19] in the $a^3\Pi$ state. The energy matrix used in the calculations [20,21] includes coupling to rotational levels up to $J = 10$ for all three Ω -ladders.

Knowing the Stark energy as a function of electric field, the force on the molecules can be calculated by taking the gradient of the Stark energy. To aid in the design of a trap, and to be able to calculate and visualize the trajectories of neutral molecules in arbitrarily shaped inhomogeneous electric fields, we slightly modified the commercially available programme SIMION [22]. The original programme calculates and visualizes the trajectories of charged particles after having determined the force acting on the particles from the gradient of the electrostatic potential. The modification to this programme is that, additional to the calculation of the electrostatic potential, the magnitude of the Stark energy of CO is calculated for each point in which the electrostatic potential is known. Subsequently, use is made of the original routine, which now calculates the force from the gradient of the Stark energy and shows the resulting trajectories.

A design for a trap based on the Stark effect, is indicated in Fig. 2. It is based on the fact that the electric field between two coaxial electrodes scales with the distance r from the axis as $1/r$, implying that molecules with a linear Stark effect, like CO for not too strong electric fields, will experience a force that scales as $1/r^2$. Stable "planetary" orbits should therefore be possible for molecules with a negative linear Stark effect in such a field geometry. To also obtain confinement in the direction along the symmetry axis, a diabolo-shaped trap is required, as depicted in Fig. 2. The figure shows both a cross section through the symmetry axis (left) and a side view (right) of the trap. The trap indicated in Fig. 2 has a diameter for the inner electrode of 2 mm, and an inner diameter of the

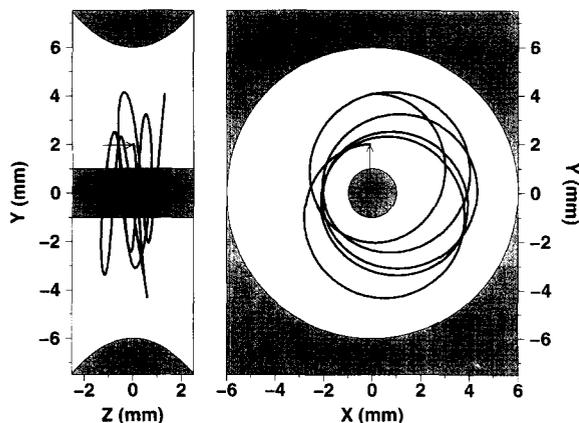


Fig. 2. Schematic view of the diabolo-shaped trap, showing a cross section of the trap in a plane through the symmetry axis (left) as well as a side view of the trap (right). The calculated trajectories of metastable CO molecules ($a^3\Pi_1$, $v = 0$, $J = 1$, $M\Omega = 1$) in the trap are indicated by the solid line in both views. The trajectory starts in the laser-excitation region, indicated with an arrow in both figures. The voltage difference applied between the inner and outer electrodes is 25 kV. The initial velocity of the molecule is 22.1 m/s in the tangential direction and 1.2 m/s in the z -direction.

outer electrode of 12 mm. The shape of the inside of the latter electrode is, in this example, parabolic. The maximum electric field inside the trap is chosen to be equal to the maximum electric field that could be realized in the above-mentioned experiments ($|E_{\max}| \approx 140$ kV/cm). This corresponds to an applied voltage difference of 25 kV between the inner and outer electrodes. When metastable CO molecules are put into this trap at the correct position and with the correct initial velocity, they will be trapped in periodic orbits. The maximum incoming velocity of the CO molecules that still allows confinement into stable periodic orbits is determined by the maximum Stark shift that can be obtained, which in turn is limited by the maximum electric field that can be achieved in practice.

A projected trajectory of a CO molecule inside the trap is indicated in both views of the trap. The trajectories are generated assuming that the CO molecules are laser prepared inside the trap in the $M\Omega = 1$ component of the $a^3\Pi_1$ ($v = 0$, $J = 1$) level using a laser beam that is running parallel to the symmetry axis and close to the inner electrode ($r = 2$ mm). Here the electric field, and hence the Stark shift, is largest. The tangential velocity of the CO molecules for which the trajectories are calculated is 22.1 m/s. Their veloc-

ity in the direction of the symmetry axis (z -axis) is 1.2 m/s. The maximum tangential velocity of the CO molecule that can be trapped is found to be 22.3 m/s for the presented configuration, assuming a maximum electric field of 140 kV/cm. The maximum velocity that can be trapped in the direction of the z -axis is found to be 2.0 m/s. It should be noted that ground-state CO molecules hardly experience any force due to electric fields because the (second-order) Stark effect in the $^1\Sigma^+$ state is negligible as compared to the Stark effect in the $a^3\Pi$ state [16].

For a practical realization of the above-mentioned scheme one can start with a slow effusive beam of CO from a liquid N_2 cooled source. In such a beam the most probable velocity is around 200 m/s, and a few percent of the molecules will have velocities below 22 m/s. After pre-selection of molecules with low velocity (using, for example, a chopper-based velocity selector), this beam of ground-state CO molecules can enter the trap perpendicularly to, and slightly above, the symmetry axis through a small (0.5 mm diameter) hole. The metastable molecules can be prepared by the above-mentioned narrow-band pulsed laser running parallel to the cylinder axis. As the electric field inside the trap scales approximately with $1/r$, there is a strong dependence of the Stark shift on the radial position. Assuming a bandwidth of 150 MHz for the preparation laser, it follows that in the radial direction only molecules over a distance of a few tens of micrometers will be in resonance with the laser. The spatial resolution of the excitation region will therefore be determined by the combination of laser bandwidth and field gradient rather than by the spot size of the laser beam. In principle, the narrow-band laser can be used for velocity selection in the z -direction by scanning through the Doppler profile. As the Stark shift is much larger than the Doppler shift, however, spatial selection rather than velocity selection will be performed.

Visualization of the trajectories of the trapped CO molecules can be performed via time- and spatially-resolved imaging of the $a^3\Pi(v = 0) \rightarrow X^1\Sigma^+(v'')$ fluorescence of the laser-prepared molecules [23]. The lifetime of the laser prepared level is around 3 ms [18], which is well matched to the typical orbiting time for CO in the trap (0.5–1.5 ms). Several round-trips are performed inside the trap during the lifetime of the metastable molecules.

CO molecules in the $a^3\Pi_2$ ($J = 2$, $M\Omega = 4$) level can be used for confinement in this geometry as well. The linear Stark effect for this quantum level is even larger than for the $a^3\Pi_1$ ($J = 1$, $M\Omega = 1$) level. The radiative lifetime of this level is around 160 ms [18] and excitation to this level is correspondingly more difficult. The much longer lifetime of this state, however, allows to molecules accumulate inside the trap for many laser pulses when using a high repetition rate preparation laser system.

Apart from confinement, electric fields can be used to manipulate polar molecules on the basis of the linear Stark effect. If, for instance, CO molecules are prepared in an electric field in a quantum level with a certain negative (positive) Stark shift, then the molecules will lose (gain) exactly this amount of kinetic energy upon entering a field-free region. The extent to which polar molecules can be slowed down or accelerated via this scheme is again determined by the magnitude of the Stark shift that can be realized.

The confinement scheme as proposed here, works for CO because these molecules can be excited inside the trap to a metastable state, where the laser is used to “switch on” the dipole moment. Otherwise, “planetary” orbits could not be accomplished. The scheme presented here will be applicable to all molecules that have this property. In addition, the scheme will also work for ground-state neutral molecules with a large dipole moment, provided the electric fields can be switched on sufficiently rapidly.

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