Chapter 7

Progress towards using CO (a^3Π) as a probe into the possible time variation of $\mu$

Abstract

A number of steps have been made to measure the two-photon transition in CO a^3Π that is highly sensitive to a variation of the proton-to-electron mass ratio. A calculation of the transition strength has shown that, in order to obtain high enough power, a resonant microwave cavity is required, which has been designed and tested. Laser excitation of the $m_J^B = 0$ sublevel is studied, and a proposal is made to improve the detection scheme.

7.1 Introduction

In this chapter, our progress towards measuring the highly sensitive two-photon microwave transition between the $\Omega = 1$, $J = 6$, + and the $\Omega = 0$, $J = 8$, + rotational levels in the a^3Π state of ^12C^16O is discussed.

In the introduction of this thesis, it was shown that a near degeneracy between three rotational levels occurs in the a^3Π state of CO. Fig. 7.1 shows the energy level diagram of these three rotational levels, using the calculations from Chapter 2. The transitions between these levels have $\Delta J = 2$, and can be induced using two-photon electric dipole transitions. In Chapter 2, it was found that the $\Omega = 1$, $J = 6$, + $\rightarrow$ $\Omega = 0$, $J = 8$, + transition, with an energy of 3.3 GHz, has a sensitivity coefficient of $K_\mu = -334$.

The proposed method to measure the two-photon microwave transition is very similar to the one-photon transition measurement discussed in Chapter 5. In this chapter, the differences will be discussed, mainly resulting from the fact that the two-photon transition requires a more intense microwave field and involves highly excited rotational levels. In section 7.2 the transition strength is
calculated and compared to the transition strength of the one-photon transition. In section 7.3 the design of a microwave cavity is presented. The obtained electric field is compared to the electric field that is required to induce the microwave transition. As the rotational levels involved have higher rotational quantum number, they have more $m^j_E$ sublevels and a lower Stark effect, the consequences of which are discussed in section 7.4. We study the effect of perpendicular $\vec{E}$ and $\vec{B}$ fields on the $\Omega = 1, J = 6, +$ state to find which $m^E_j$ sublevel needs to be populated, and which UV transition can be used to populate it. Furthermore, we will discuss the lower separation of the initial and final states on the MCP detector due to the lower Stark shift.

**Figure 7.1:** The relevant rotational levels of the energy level diagram of the $a^3\Pi$ state of $^{12}\text{C}^{16}\text{O}$. The three nearly degenerate rotational levels are shown, with the rotational quantum number and the total parity indicated.
7.2 Transition strength

As the highly sensitive transition is a two-photon transition, it requires a more intense microwave field than the one-photon transition measured in Chapter 5. Here, we present an analysis of the transition strength of the transition between the $\Omega = 1, J = 6, +$ level and the $\Omega = 0, J = 8, +$ level.1

The transition strength is given by:

$$\frac{\Omega_{Rabi}}{2\pi} = \sum_j \left\{ \left( \frac{1}{2} \right)^2 \cdot |E|^2 \cdot \mu_e^2 \right\}$$

$$\left( \frac{E_j - \frac{1}{2} (E' + E'')}{(E_j - \frac{1}{2} (E' + E''))} \right)$$

$$\times \left( J' + 1 \right) \sum_i g(\Omega_i, J') \times c_{\Omega_i,\Omega',J'} \times c_{\Omega_i,\Omega_j,J_j}$$

$$\times \left( J_j + 1 \right) \sum_i g(\Omega_i, J_j) \times c_{\Omega_i,\Omega_j,J_j} \times c_{\Omega_i,\Omega'',J''} \right\}.$$  (7.1)

Here, the summation over $j$ is over all intermediate states. In this case, there are three, the $J = 7, -$ level in all three spin-orbit manifolds. However, the $\Omega = 2, J = 7, -$ level couples weakly to both initial and final state, so this intermediate state is neglected. In this calculation, the Hamiltonian matrix and molecular constants found in Chapter 2 are used. The summation over $i$ is over all three values of $\Omega$, single primes indicate the initial state, double primes the final state. The $c$-coefficients are the squares of the eigenvectors of the Hamiltonian matrix, representing the amount of $\Omega_i$ character a level has. $|E|$ is the absolute value of the electric field and $\mu_e$ is the electric dipole moment of the molecule. $E$ is the energy of the corresponding rotational level. $g(\Omega, J)$ is the HönL-London factor of the transition, for $\Delta m = 0$ transitions, and is given by:

$$g(\Omega, J) = \frac{1}{J+1} \sqrt{\frac{(J+1)^2 - \Omega^2}{(2J+1)(2J+3)}}.$$  (7.2)

This leads to a transition strength of $T = 2.6 \times 10^{-3} (\mu_e E)^2$ Hz, compared to $0.1 \mu_e E$ Hz for the one-photon transition measured in Chapter 5, both with $E$ in V/cm and $\mu_e$ in Debye. This transitions strength comes from two different intermediate levels, the $\Omega = 0, J = 7, -$ and $\Omega = 1, J = 7, -$ levels. Of these two, the coupling via the $\Omega = 0, J = 7, -$ level is approximately two times stronger than the coupling via the $\Omega = 1, J = 7, -$ level, but both couplings are opposite in sign.

1This calculation was performed by Dr. W.L. Meerts (Radboud University Nijmegen, the Netherlands).
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7.3 Microwave cavity

To enhance the strength of the applied microwave field, a resonant cavity is used. In this section, we will discuss the design and testing of such a cavity.

For the shape of the cavity we choose a cylindrical hole in a copper block, commonly known as a pillbox cavity\(^2\). A schematic drawing is shown in Fig. 7.2. We drive a Transverse Magnetic (TM) mode, meaning that the electric field of the microwave field is along the $z$-axis, parallel to the bias magnetic field, driving $\Delta m_B^J = 0$ transitions. For the TM modes, the resonance frequency of a pillbox cavity is given by:

$$\omega_{mnp} = c \sqrt{x_{mn}^2 / R^2 + p^2 \pi^2 d^2}.$$  \hfill (7.3)

where $x_{mn}$ is the $n$-th root of $J_m(x) = 0$. The mode numbers $m$, $n$, $p$ are the mode number in the $\phi$-, $r$-, and $z$-directions, respectively. For the lowest order, and smallest, TM mode, the TM$_{010}$ mode, this equation simplifies to:

$$R_{cav} = \frac{cx_{01}}{2\omega_0} = \frac{2.405c}{2\pi f_0}.$$  \hfill (7.4)

For a frequency of 1.6283 GHz, this gives a radius of 70.47 mm, while the height of the cavity is not constrained by the resonant mode, and can thus be chosen freely. We have chosen a height of 50 mm.

The design of the cavity can be seen in Fig. 7.3. It is made out of three oxygen-free copper parts, one body and two 'lids', and four KF-flanges. The molecules pass through the flanges in the lids, while the other two flanges are

\(^2\)We based our design and calculations on the work of van Oudheusden [149].
used for the microwave input and a plunger, that can be used to tune the cavity resonance. The plunger is a 15 mm diameter copper cylinder, attached to a micrometer screw. It can be moved in and out of the cavity over a distance of \(\sim 15\) mm, thereby affecting the field and shifting the resonance frequency. The resonance frequency of the cavity is measured using a directional coupler to split of the microwave signal that is reflected back from the cavity. This back-reflected signal is fed into a microwave power meter, and the output of the power meter is recorded as a function of frequency. Typical measurements can be seen in Fig. 7.4, for three different settings of the plunger. The frequency of the microwave radiation is scanned, and when the input is resonant with the cavity a large fraction of the power is coupled into the cavity. Therefore the reflected power drops. From the figure we can see that the resonance frequency can be tuned over approximately 4 MHz. The frequency of the transition is known from the UV-measurements to be 1628.3(5) MHz, well within the tunable range of the cavity. To further maximize the intra-cavity power, the impedance of the cavity needs to be matched to the cable. This is done by rotating the antenna. From Fig. 7.4, it can be seen that more than 95% of the input microwave power can be coupled into the cavity. The Q-factor of the cavity is given by the ratio between the central frequency of the resonance and the width of the resonance, \(Q = \nu/\Delta \nu = 1628/0.28 = 5.8 \times 10^3\).

Now that we know the electric-field build-up in the cavity, we can calculate
how much microwave power we need to input into the cavity to be able to drive the two-photon transition. We first write the required electric field

$$E_{\text{req}}^2 = \frac{2 \arcsin(\sqrt{p})}{2\pi\tau T \mu_E^2},$$

(7.5)

with $p$ the requested probability that a molecule makes a transition, $T$ as following from Eq. 7.1 and $\tau$ the time it takes molecules to fly through the microwave field. To calculate the electric field that is present in the microwave cavity, we use

$$E_{\text{cav}}^2 = \frac{2PQ}{Ac\varepsilon_0},$$

(7.6)

with $P$ the microwave power input into the cavity, $Q$ the quality factor as above and $A$ the surface area of the cavity. We need the electric field in the cavity to be equal or higher than the electric field required to drive the transition. To calculate how much input power we need, we write

$$P_{\text{req}} = \frac{E_{\text{req}}^2 Ac\varepsilon_0}{2Q}.$$  

(7.7)

To find the required amount of input microwave power we use $\tau = l/v = 0.05/800 = 62.5 \times 10^{-6}$ s, $A = \pi R_{\text{cav}}^2 = \pi 0.0704^2 = 1.6 \times 10^{-2}$ m$^2$ as given by the cavity designed above and $\mu_e = 1.37$ D. For the 1.63 GHz transition, we find $E_{\text{req}} = 911$ kV/cm for a transition probability of 0.5. We now find that that $P > 30$ W.

### 7.4 Laser excitation

In the one-photon lambda-doublet transition measurement discussed in Chapter 5, the Zeeman shift caused by the bias magnetic field was canceled by averaging over two microwave transitions with opposite Zeeman shift, as the $m_B^J = 0 \rightarrow m_B^J = 0$ transition was not allowed. As a result, instability of the bias magnetic field was the largest contributing error. In the two-photon transition discussed here, the $m_B^J = 0 \rightarrow m_B^J = 0$ transition is allowed, greatly limiting the effect of magnetic fields on the measurement. In Chapter 5, the correspondence between $m_B^J$ and $m_E^J$ sublevels was studied. For the $J = 6$ level, 13 sublevels are present. The splitting of the ground state is much smaller than the laser bandwidth, and can thus be ignored. Meek et al. [128] describe the $\Omega = 1, J = 1$ level of CO $a^3\Pi$ in perpendicular $\vec{E}$ and $\vec{B}$ fields. Here, we follow their description, and extend it to higher $J$ levels. We choose the magnetic-field vector along the $z$-axis and the electric-field vector in the $xy$-plane. States are labeled by the quantum number $m_B^J$, the projection of the total angular momentum on the $z$-axis. As $^{12}\text{C}^{16}\text{O}$ has zero nuclear spin, total angular momentum is given by $J$ only. The magnetic field is parallel to the quantization axis, and therefore only couples sublevels with the same $m_B^J$ and parity. The
electric field only couples sublevels with $\Delta m_j^B = \pm 1$ and opposite parity. We can now write the Hamiltonian matrices, one for each parity, using the matrix elements given by

$$
\langle \Lambda S \Sigma J \Omega m | H_{\text{stark}} | \Lambda \Sigma \Sigma' J' \Omega' m' \rangle = \\
\mu_e E (-1)^{m-\Omega+1} \sqrt{(2J+1)(2J'+1)} \\
\begin{pmatrix} J & 1 & J' \\ -m & -1 & m' \end{pmatrix} \begin{pmatrix} J & 1 & J' \\ -\Omega & 0 & \Omega \end{pmatrix}
$$

(7.8)

and

$$
\langle \Lambda S \Sigma J \Omega m | H_{\text{zeeman}} | \Lambda \Sigma \Sigma' J' \Omega' m' \rangle = \\
\mu_b B (-1)^{m-\Omega} \sqrt{(2J+1)(2J'+1)} \\
\begin{pmatrix} J & 1 & J' \\ -m & 0 & m' \end{pmatrix} \begin{pmatrix} J & 1 & J' \\ -\Omega & \Omega - \Omega' & \Omega' \end{pmatrix}
$$

(7.9)
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**Figure 7.5:** Dependence of different \(m_J\) sublevels on the electric field in a perpendicular magnetic field of 50 Gauss.

for the electric and magnetic-field coupling, respectively. Here, \(\mu_e\) and \(\mu_b\) are the electric and magnetic dipole moments, \(E\) and \(B\) are the absolute values of the electric and magnetic fields, respectively.

Fig. 7.5 shows the energy shifts in combined, perpendicular \(\vec{E}\) and \(\vec{B}\) fields for the \(\Omega = 1, J = 6, +\) level as a function of the electric field in a fixed magnetic field of 50 Gauss. As two \(m_J\) sublevels are degenerate in an electric field, two different \(m_J^B\) sublevels combine to form one \(m_J^E\) sublevel, where \(m_J^E\) is the projection of the total angular momentum on the electric field vector. In the experiment, we need to laser excite the \(m_J^B = 0\) sublevel. Therefore, we need to adiabatically follow the \(m_J^B = 0\) sublevel to high electric field, and see that it becomes the \(m_J^E = 3\) sublevel. This sublevel can be reached from two ground state rotational levels, via the \(R_2(5)\) and the \(P_2(7)\) transitions. As the beam of CO molecules is rotationally cold, we would prefer to excite from as low a ground state rotational level as possible, and thus use the \(R_2(5)\) transition. Unfortunately, this transition is overlapped with the \(R_2(7)\) transition, as shown in Fig. 7.6, where a calculation of both transitions is plotted in an electric field of 15 kV/cm and a rotational temperature of 20 K. Note, that in this figure the intensity of the \(R_2(7)\) transition is multiplied by a factor 25 for clarity. The \(R_2(5)\) transition to the \(m_J^E = 3\) sublevel for this electric field strength is in
Figure 7.6: A simulation using pgopher of the $R_{2}(5)$ and $R_{2}(7)$ transitions in an electric field of 15 kV/cm, with a rotational temperature of 20 K. The $R_{2}(7)$ transition is multiplied by a factor of 25 for clarity.

between the $R_{2}(5)$ transition to the $m_{E}^{F}=2$ sublevel and the $R_{2}(7)$ transition to the $m_{E}^{F}=0$ sublevel. Exciting in a higher electric field will give more energy separation between the different sublevels of the $R_{2}(5)$ transition, but starting from around 20 kV/cm the $R_{2}(7)$ transition will be excited as well, adding a small background to the undeflected signal.

As discussed, the $m_{B}^{J}=0$ sublevel becomes the $m_{E}^{J}=3$ sublevel. However, populating the $m_{E}^{F}=3$ sublevel will also populate the $m_{B}^{J}=-1$ sublevel. Molecules in this sublevel will not make a microwave transition, therefore the population of the initial state will not be empty at the resonance of the microwave transition. This problem was circumvented in the one-photon measurement by exciting the $m_{E}^{F}=0$ sublevel, which only leads to one $m_{B}^{J}$ sublevel.

7.5 Current status and outlook

Microwave frequency scans have been performed to observe the two-photon transition, at two different occasions during the project, but they were ultimately unsuccessful. In the first run, a number of scans were done with the resonant cavity in place, but it was found that the observed spatial separation
Progress towards using CO ($a^3Π$) as a probe into drifting $\mu$ between the different $m^E_J$ sublevels was insufficient; the depletion of the signal of the $m^E_J = 6$ sublevel induced by the microwave transition would be hidden in the background. In order to obtain a larger deflection, a new deflection field was constructed. This deflection field has a length of 60 cm instead of 30 cm and it was conditioned to 18 kV instead of 12 kV. These factors combined led to a factor 4 improvement of the observed deflection. Preliminary measurements were taken with the new deflection field, however, around this time calculations became available for the transition strength of the two-photon transition and it became clear that the microwave power delivered by our generator (200 mW) was smaller than required to make a Rabi flop (30 W), and a microwave amplifier was acquired. Due to time limitations, this amplifier was never used to search for the two-photon transition.

Even though it should be possible to observe the two-photon transition with the new deflection field and microwave amplifier, the number of detected molecules in the current experiment is probably too small to perform a competitive test of the time-variation of $\mu$. The main reason for this is the small population of the $J = 5$ level of the ground state in the molecular beam. Improved signal on the $R_2(5)$ transition was achieved by lowering the pressure behind the valve from 2 bar to 0.5 bar, resulting in less rotational cooling in the expansion. The downside to this approach is that by raising the rotational temperature of the beam the $R_2(7)$ transition becomes stronger relative to the $R_2(5)$ transition. This leads to an increased background. Another reason for low signal is the small Stark shift of the $J = 6$ level which makes it necessary to use a long deflection field. The longer flight path leads to geometrical losses due to the divergence of the molecular beam scaling with the square of the length of flight. Furthermore, as a result of the longer flight path, a larger fraction of the molecules will decay back to the ground state before reaching the detector.

The signal could be improved in two ways. The geometric losses can be reduced by focusing the beam of molecules using quadrupole or hexapole lenses [54]. However, due to the low effective electric dipole moment of the involved states, this will require long lenses at a very high voltages. Another option to improve the signal would be to improve the detection efficiency. In the current experiments, the molecules are detected by letting the metastable molecules impinge on an MCP. This process has a quantum efficiency of only $\sim 10^{-3}$ [54], as the work function of the surface of the MCP is only slightly smaller than the 6 eV of internal energy of the metastable molecules. Increasing the efficiency of the detection would greatly improve the accuracy of the experiment, and could be achieved by letting the metastable molecules impinge on a gold surface, see Fig. 7.7 for a schematic overview. The deflected and undeflected beams of metastable molecules each impinge on a separate gold plate, freeing electrons that are collected by a high voltage grid and then detected. To further improve the detection, the gold plate could be heated, to anneal it in between measurements, or a sodium beam could be installed, to constantly
deposite a layer of sodium atoms on the surface. This would further lower the work function of the surface and keep the surface free from contamination.

To estimate the signal strength that is required for a competitive measurement, we use the Allan deviation, given by

$$\sigma(\tau) = \frac{1}{Q\sqrt{N\tau/\tau_c}},$$  \hspace{1cm} (7.10)

with $\sigma(\tau)$ the final error on the measurement, $Q$ the quality factor of the measurement, given by the transition frequency over the measured linewidth, $\nu/\Delta \nu$, $N$ the amount of molecules that is detected per measurement cycle, $\tau$ the total measurement time and $\tau_c$ the time of one measurement cycle [150]. The quality factor that can be reached, as shown by the one photon measurement, is $1.6 \times 10^9/1.4 \times 10^3 \approx 1.1 \times 10^6$. From previous experiments we expect that replacing the mcp by the new detector will increase the number of detected molecules with a factor 100. Together with other improvements it should be possible to detect 1000 molecules per shot. The current best constraint on $\mu$
variation is set by the SF$_6$ experiment of Shelkovnikov [35], with an accuracy of $5.6 \times 10^{-14}$. To be competitive with that measurement, an accuracy of $1.8 \times 10^{-11}$ is required, as our planned experiment has a sensitivity coefficient of 330. This leads to a total measurement time of $\sim 60$ hours. This long measurement time makes searching for systematic effects challenging, but not impossible.

7.6 Conclusion

To measure the $\Omega = 1, J = 6, + \rightarrow \Omega = 0, J = 8, +$ transition, a resonant cavity has been designed and tested. The deflection has been enhanced by making a longer deflection field, that has been conditioned to higher voltages. Due to time limitations, we have not been able to search for the two-photon transition with the acquired microwave amplifier. Although it should be possible to detect the two-photon transitions in the present setup, it will be challenging to reach the necessary accuracy to improve the current limit on a possible time variation of the proton-to-electron mass ratio. For a competitive test it is necessary to implement a more sensitive detector and/or to use quadrupole or hexapole lenses to collimate the beam.
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