Supplementary Material for "Laser Cooling Scheme for the Carbon Dimer ($^{12}C_2$)"

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CALCULATION OF BRANCHING RATIOS

After identifying three excited states for possible cycling schemes as described in the main text, we identified their decay paths using the ExoMol database [1–4] and compiled the respective transition energies and Einstein A coefficients, A_i. The subscript i identifies each decay path. In order to determine the relative strength and thus the relative importance of the decay paths, we calculate the branching ratios (BRs) via

$$\mathrm{BR}_i = \frac{A_i}{\sum_j A_j}, \tag{1}$$

where j runs over all decay paths. Branching ratios are such that

$$\sum_j A_j = \Gamma, \tag{2}$$

where Γ is the total decay rate of the excited state, i.e., $\Gamma = \tau^{-1}$, where τ is the lifetime of the excited state. Figure 3 of the main text plots the decay BRs for all excited states vs. transition wavelength λ_i. Decay paths with BRs $< 10^{-6}$ are deemed negligible for our considerations and omitted from the calculations and the figure.

CLOSURE OF THE CYCLING SCHEMES

In order to quantify the level of closure of the cycling schemes, we determine how many scatterings can be performed for a given number of repump lasers addressing the decay paths until 10 % of molecules remain in cycling-states, that is until 90 % of the molecules are lost to dark states. In addition, we determine the time necessary to complete those scatterings.

We model photon scattering as a Bernoulli process with a probability p that the molecule will remain within the cycling scheme per photon scattering [5] and a probability $1-p$ that the molecule will leave. The quantity p is given by

$$p = \sum_i \mathrm{BR}_i, \tag{3}$$

where i runs over all driven transitions (the main cooling transition and the addressed repump transitions). The fraction of molecules in bright states after n photon scatterings is given by p^n. The number of scatterings that retain 10% of the molecules in a bright state is

$$n_{10\%} = \ln(0.1)/\ln(p). \tag{4}$$

The time to complete $n_{10\%}$ scatterings is given by

$$t_{10\%} = n_{10\%}/R, \tag{5}$$

where R denotes the scattering rate. The time is related to the natural linewidth of the excited state via

$$R = \Gamma \cdot n_e, \tag{6}$$

where n_e is the probability that the molecule occupies the excited state during the photon cycling process [6]. This takes into account saturation effects and the number of transitions addressed and is given by

$$n_e = \frac{1}{G + 1 + 2\sum I_{\mathrm{sat},i}/I_i}, \tag{7}$$

as discussed in Ref. [7]. Here, I_i is the intensity of the laser addressing the i^{th} transition; $I_{\mathrm{sat},i} = \pi hc\Gamma/3\lambda_i^3$ is the associated saturation intensity [6]; G is the number of driven transitions; h is Planck's constant; and c is the speed of light.

We also note that the definition of saturation intensity for a particular transition, $I_{\mathrm{sat},i}$, between the excited state and one of the lower states in a multi-level system (as relevant here) is identical to the saturation intensity for a two-level system with a scattering rate given by the transition's Einstein A_i coefficient, weighted by the inverse probability that such a transition will occur (given by the inverse of its branching ration), that is

$$I_{\mathrm{sat},i} = \frac{\pi hcA_i}{3\lambda_i^3} \cdot BR^{-1} = \frac{\pi hcA_i}{3\lambda_i^3} \cdot \frac{\sum_j A_j}{A_i}. \tag{8}$$

The weighing by the inverse branching ratio is done so that a transition that is 100 times weaker than the sum of the other ones (i.e., BR = 0.01) will need 100 times more intensity to get saturated.

MOLECULE DEFLECTION

For deflection, molecules scatter photons and experience a recoil that leads to an increasing Doppler shift δ as more photons are scattered. We assume that molecules continue scattering photons until the Doppler shift is equal to

$$\delta = \Gamma/2\sqrt{1 + I/I_{\mathrm{sat}}}. \tag{9}$$

Given that the Doppler shift is $\delta = v \cdot k = v \cdot \frac{2\pi}{\lambda}$, where v is the molecule velocity and k is the light wave number, the velocity at which the molecule is tuned out of resonance is given by

$$v_\delta = \frac{\Gamma\lambda\sqrt{1 + I/I_{sat}}}{4\pi}. \tag{10}$$

To find the number of scatterings n_{defl} that lead to a recoil velocity v_δ, we divide the momentum associated with this velocity by the photon momentum $p = \hbar k$, with $\hbar = h/2\pi$. If we only consider the main cooling transition, this yields

$$n_{\mathrm{defl}} = \frac{m_{\mathrm{C_2}} v_\delta}{p}, \tag{11}$$

where $m_{\mathrm{C_2}}$ is the mass of a carbon dimer. When repump transitions are addressed, we calculate v_δ using the strongest transition, i.e. the transition with largest BR. For the photon momentum, we use the mean value

$$\bar{p} = \sum_j \hbar k_j \cdot \frac{BR_j}{\sum_i BR_i}, \tag{12}$$

which weighs the photon momentum of each transition $\hbar k_j$ by the probability of a scattering on transition j to happen. Here, i and j run over the addressed transitions. L_{defl}, the axial distance travelled by the molecules until they leave resonance, is given by the product of the initial velocity, v_0, and the time to complete n_{defl} scatterings, $t_{\mathrm{defl}} = n_{\mathrm{defl}}/R$, so that $L_{\mathrm{defl}} = v_0 \cdot t_{\mathrm{defl}}$. The deflection angle is calculated via

$$\theta = \arctan\left(\frac{v_\delta}{v_0}\right). \tag{13}$$

COOLING

For cooling, we find the number of scatterings n_{cool} by dividing the initial molecule momentum by the mean photon momentum $\bar{p}$. Then, we determine the cooling time t_{cool} from the scattering rate and the number of photon scatterings via $t_{\mathrm{cool}} = n_{\mathrm{cool}}/R$ and the length required for cooling from $L_{\mathrm{cool}} = v_0 \cdot t_{\mathrm{cool}}/2$. The reported accelerations are calculated via $a = \frac{\hbar k R}{m_{\mathrm{C_2}}}$.

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