

# ThO Neon Beam Flux Calculation

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We will calculate the peak and total flux (per pulse) of a 17K neon-cooled ThO beam. The calculation will use data from the run on 24 January 2010, shown below.

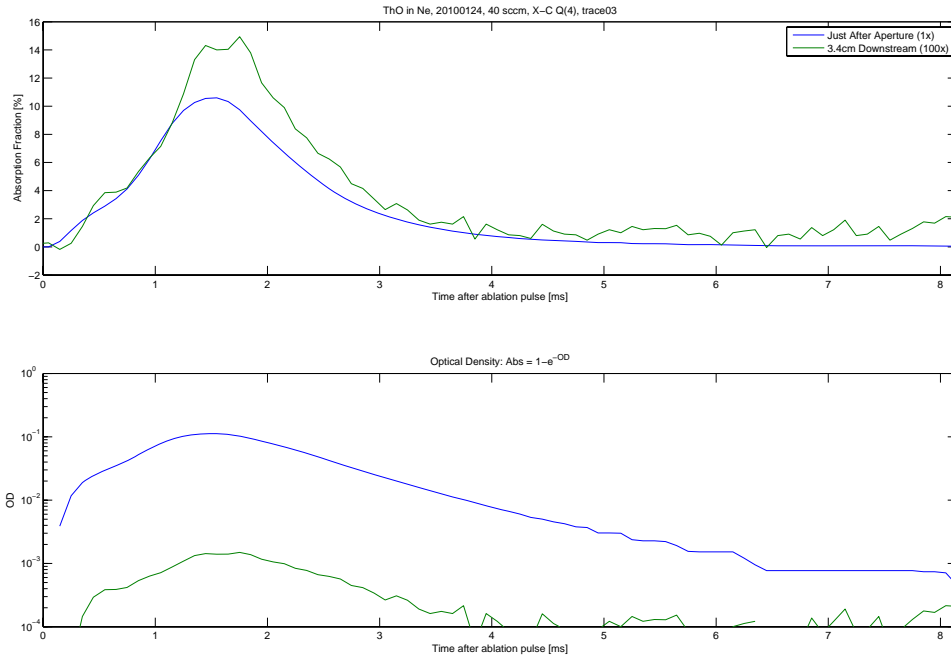


Figure 1: Typical absorption data on the ThO X-C Q(4) line. The blue curve is absorption just after the cell aperture, while the green curve is absorption 3.4cm after the aperture. The green absorption fraction curve is scaled up by a factor of 100. There is a 2mm diameter collimating aperture 1.2cm downstream from the cell. The cell temperature is 17K, and there is a 40 sccm flow outside of the aperture, which is 4 mm by 4 mm square.

The important values from this data are:

$$\boxed{OD_{\text{peak}} \approx 0.1 \quad \int dt OD(t) \approx 0.2 \times 10^{-3} \text{ s}}$$

The cell aperture is 5.4mm diagonal, or 3.8mm x 3.8mm. We wish to extract the number density  $n$  at the aperture by solving  $n \approx 1/(\ell\sigma)$ , where  $\sigma$  is the absorption cross section and  $\ell$  is the optical path length. Since we are sending the laser along the diagonal of the aperture, the path length is  $\ell \approx 5\text{mm}$ .

The resonant  $|g, J\rangle \rightarrow |e, J'\rangle$  absorption cross-section for a doppler-broadened atom in zero field is given by

$$\sigma_{abs} = \frac{\sqrt{\pi} \lambda^2 2J' + 1 \Gamma_p}{2 2\pi 2J + 1 \Gamma_D}$$

where  $\Gamma_D$  is the doppler width of the excited state, and  $\Gamma_p$  is the width of the decay  $|e, J'\rangle \rightsquigarrow |g, J\rangle$  (see Dave's book, §3.5 and 3.6).

The natural width of an electronic transition is  $\gamma \sim 2\pi \times 10$  MHz, so we would require temperatures of  $\sim 1$  K in ThO for the Doppler width to be comparable to the natural width. Thus we are indeed in the doppler broadened limit. This  $\gamma$  corresponds to a natural lifetime of about 15 ns, so it is definitely a lower bound.

In the appendix, we show that  $\Gamma_p \approx \frac{3}{4}\gamma$ . Therefore, for a Q-branch ( $\Delta J = 0$ ) absorption transition from  $|X, v = 0\rangle \rightarrow |C, v' = 0\rangle$ ,

$$\begin{aligned} \sigma &= \frac{\sqrt{\pi} \lambda_{XC}^2 \frac{3}{4}\gamma}{2 2\pi \Gamma_D} \\ &= \frac{\sqrt{\pi} (690 \text{ nm})^2 \frac{3}{4} \times 2\pi \times 10\text{MHz}}{2 2\pi 2\pi \times 50\text{MHz}} \\ &\approx 1 \times 10^{-10} \text{cm}^2 \end{aligned}$$

where we used that  $\Gamma_D = 2\pi \times \sqrt{2K_B T/m\lambda^2}$ , and we have assumed that the molecules at the aperture are at 17K. Therefore the peak density outside the cell is

$$n_{\text{peak}, J=4} \approx \frac{1}{\ell \sigma} = \frac{1}{(5 \text{ mm})(1 \times 10^{-10} \text{cm}^2)} \approx 2 \times 10^{10} \text{cm}^{-3}$$

Now let's try to extract the total number of molecules in this state per pulse, and the peak flux. If the density outside the aperture is  $n(t)$ , the aperture area is  $A$ , and the molecules are moving forward at velocity  $v$ , then the instantaneous molecular flux is

$$\dot{N}_{J=4}(t) = Avn(t) \approx \frac{Av}{\ell \sigma} OD(t) = \frac{(4 \text{ mm})^2 (150 \text{ m/s})}{(5 \text{ mm})(1 \times 10^{-10} \text{cm}^2)} OD(t) = (5 \times 10^{13} \text{ s}^{-1}) OD(t)$$

Where we have estimated  $v \approx 150$  m/s. The peak OD is 0.1, so the peak flux is

$$\dot{N}_{\text{peak}, J=4} \approx 5 \times 10^{12} \text{ s}^{-1}$$

The total number of molecules per pulse is then

$$N_{\text{tot}, J=4} \approx (5 \times 10^{13} \text{ s}^{-1}) \int dt OD(t) = (5 \times 10^{13} \text{ s}^{-1})(0.2 \times 10^{-3} \text{ s}) = 1 \times 10^{10}$$

Remember this is the number of molecules in the  $J = 4$  state at 17K, which should contain 14% of the total number of molecules. We expect the beam to cool as it expands, say to 4K. At this temperature the  $J = 1$  state has about 25% of the total population, so we can estimate that the  $J = 1$  population in the beam will be increased by a factor of  $25/14 \approx 2$ , for a total number per state per pulse of  $\approx 2 \times 10^{10}$ .

## Appendix: Branching Ratios

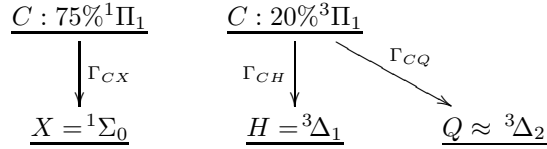
In a molecule the partial width is the total spontaneous decay rate of the excited state multiplied by the electronic, vibrational (Franck-Condon), and rotational (Hönl-London) branching ratios into the lower state. Denote these branching ratios as  $f_e, f_v, f_r$  respectively. The partial width of a Q-branch, X-C transition is approximately 3/4 the total width, as we will now show by calculating each of these branching ratios.

### Electronic

The C state can undergo an E1 decay into the states X,H,Q,A, or B. The decay rates are proportional to the wavelength of the transition cubed, and notice

$$\log \lambda_{CX}^3 = 8.3 \quad \log \lambda_{CH}^3 = 9.1 \quad \log \lambda_{CQ}^3 = 9.2 \quad \log \lambda_{CA}^3 = 10.2 \quad \log \lambda_{CB}^3 = 10.4$$

We can therefore estimate that decays to H and Q are a  $\sim 10\%$  effect, while the others are  $\sim 1\%$  and can be ignored. The majority of the C state is a combination of  $75\%{}^1\Pi_1 + 20\%{}^3\Pi_1$ . Since the X and Q/H states are singlet and triplet respectively, the strongest transitions will be from the singlet-singlet and triplet-triplet decays, i.e.



The branching ratios are then

$$\begin{aligned}
 \frac{\Gamma_{CH}}{\Gamma_{CX}} &= \frac{20\% \omega_{CH}^3 |\mathbb{D}[{}^1\Pi_1 \rightarrow {}^0\Sigma_1]|^2}{75\% \omega_{CX}^3 |\mathbb{D}[{}^3\Pi_1 \rightarrow {}^3\Delta_1]|^2} \\
 &\approx \frac{20\% (1/1090)^3}{75\% (1/690)^3} \\
 &\simeq 7\% \\
 \frac{\Gamma_{CQ}}{\Gamma_{CX}} &= \frac{20\% \omega_{CQ}^3 |\mathbb{D}[{}^1\Pi_1 \rightarrow {}^0\Sigma_1]|^2}{75\% \omega_{CX}^3 |\mathbb{D}[{}^3\Pi_1 \rightarrow {}^3\Delta_2]|^2} \\
 &\approx \frac{20\% (1/1195)^3}{75\% (1/690)^3} \\
 &\simeq 5\%
 \end{aligned}$$

where  $\mathbb{D}$  represents the electronic dipole matrix element between the pure  ${}^{2\Sigma+1}\Lambda_\Omega$  states. Therefore the electronic branching ratio back down to X is

$$\frac{\Gamma_{CX}}{\Gamma_{CX} + \Gamma_{CH} + \Gamma_{CQ}} = \frac{1}{1 + \Gamma_{CH}/\Gamma_{CX} + \Gamma_{CQ}/\Gamma_{CX}} = 89\%$$

$$\boxed{X - C \text{ Electronic Branching Ratio } f_e \approx 0.9}$$

## Vibrational

The probability of a state  $|\text{Non-vibrational, Vibrational}\rangle = |\Gamma_1, i\rangle$  decaying into state  $|\Gamma_2, f\rangle$  is proportional to  $F_{if} \times M[\Gamma_1, \Gamma_2]$ , where  $M$  is some matrix element that doesn't depend on the vibrational wavefunctions, and  $F_{if}$  is the Franck-Condon factor, or overlap of the vibrational wavefunctions:

$$F[1 \rightarrow 2]_{if} = |\langle f|i\rangle|^2 = \left| \int dx \langle x|f\rangle^* \langle x|i\rangle \right|^2$$

Therefore, the factors  $F_{if}$  give the vibrational branching ratios for a particular electronic decay. The  $v^{\text{th}}$  vibrational wavefunction for a simple harmonic oscillator with reduced mass  $m$ , natural frequency  $\omega$ , and equilibrium position  $r$  is given by

$$\psi_v(x) = \frac{1}{\sqrt{2^v(v!)}} \left( \frac{m\omega}{\pi\hbar} \right)^{1/4} H_v((x-r)\sqrt{m\omega/\hbar}) e^{-m\omega(x-r)^2/(2\hbar)}$$

where  $H_v$  is a Hermite polynomial. The Franck-Condon factor between the  $i$  and  $f$  vibrational levels are then

$$F_{if} = \frac{m\sqrt{\omega_i\omega_f}}{2^{(i+f)}i!f!\pi\hbar} \left| \int dx H_i((x-r_i)\sqrt{m\omega_i/\hbar}) H_f((x-r_f)\sqrt{m\omega_f/\hbar}) e^{-m[\omega_i(x-r_i)^2 + \omega_f(x-r_f)^2]/(2\hbar)} \right|^2$$

The  $X$  and  $C$  states have the following molecular constants:

State	$\omega[cm^{-1}]$	$r[\text{\AA}]$
$X$	895.77	1.83
$C$	825.1	1.87

Plugging these values into a simple computer program shows that

$$F[C \rightarrow X]_{0f} = \{83, 16, 1, 0, \dots\}\%$$

Where we have rounded to the nearest percent. Therefore, when the the  $|C, v=0\rangle$  decays into the  $X$  state, 83% of the time it will end up in  $|X, v=0\rangle$ .

The above calculation assumes that the vibrational potential is a harmonic oscillator; a more accurate calculation is to treat the potential as a Morse potential. Luckily somebody has done this for us<sup>1</sup>, and their 0-0 branching ratio for  $X - C$  is 0.86. Therefore, after all of the stuff up above,

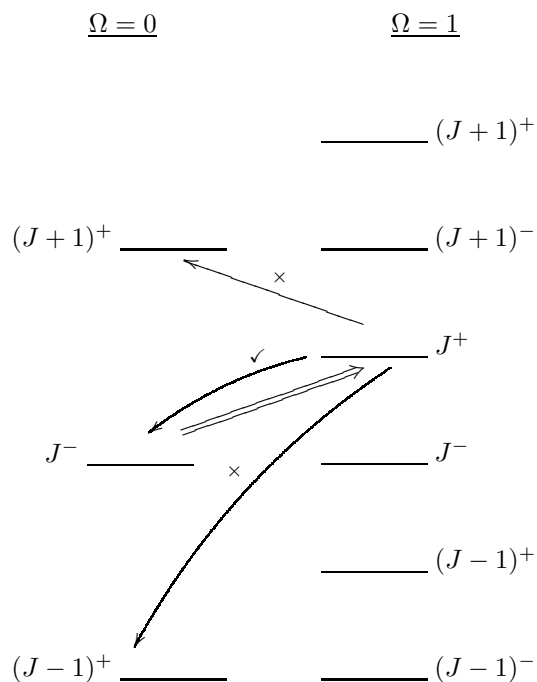
$X(v=0) - C(v=0) \text{ Vibrational Branching Ratio } f_v \approx 0.86$

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<sup>1</sup>Wentink, T., & Spindler, R. J. (1972). J. Quant. Spectrosc. and Rad. Transf., 12(11), 1569.

## Rotational

Now assume that we are looking at a  $Q$ -branch line, i.e.  $J = J'$ . Further assume that we are looking at states with  $\Omega = 0, \Omega' = 1$ , for example  $X - C$  absorption. Then the state  $|J, \Omega' = 1\rangle$  can only decay back to  $|J, \Omega = 0\rangle$  by parity, as seen in the following figure.



The figure above shows the level structure (not to scale). The  $\Omega = 1$  state has  $\Omega$  doublets of opposite parity for each  $J$ , whereas the  $\Omega = 0$  state does not. Thus if the bold line  $\Rightarrow$  indicates the absorption transition, we can see that the only E1 allowed decay ( $+ \leftrightarrow -$  parity,  $\Delta J = 0, \pm 1$ ) is back down to  $J$  in the lower state. Therefore the rotational branching ratio for a  $Q$  branch transition is 1.

$$\boxed{X - C \text{ } Q - \text{branch Electronic Branching Ratio } f_r = 1}$$

## Overall Branching Ratio

The conclusion is that the branching ratio of the decay  $|C, v = 0, J\rangle \rightsquigarrow |X, v = 0, J\rangle$  is approximately

$$f_e \times f_v \times f_r = (0.9) \times (0.86) \times (1) = \boxed{0.77 \approx \frac{3}{4}}$$