June 19, 2021

Iterative Method of Calculation

The basic method of calculating the tuneout frequency $\omega_{\rm TO}$ is to expand the various contributions to the frequency-dependent polarizability $\alpha(\omega)$ in a Taylor series about a starting value ω_i in the form

$$\alpha(\omega) = \alpha_{\rm NR}(\omega_i) + \left.\frac{d\alpha_{\rm NR}(\omega)}{d\omega}\right|_{\omega_i}(\omega-\omega_i) + \frac{1}{2}\left.\frac{d^2\alpha_{\rm NR}(\omega)}{d\omega^2}\right|_{\omega_i}(\omega-\omega_i)^2 + \Delta\alpha(\omega_i) + \left.\frac{d\Delta\alpha(\omega)}{d\omega}\right|_{\omega_i}(\omega-\omega_i) \quad (1)$$

where $\alpha_{\rm NR}(\omega)$ is the nonrelativistic polarizability, $\Delta\alpha(\omega)$ denotes the other relativistic corrections (but excluding for now the QED and other corrections), and $i = 0, 1, 2, \cdots$ for a sequence of iterations to convergence. The sequence of steps is then as follows:

1. Find a starting value ω_0 for which the nonrelativistic polarizability satisfies $\alpha_{\rm NR}(\omega_0) = 0$. This defines the nonrelativistic tuneout frequency.

2. Solve the quadratic equation

$$\alpha_{\rm NR}(\omega_0) + \left.\frac{d\alpha_{\rm NR}(\omega)}{d\omega}\right|_{\omega_0}(\omega-\omega_0) + \frac{1}{2}\left.\frac{d^2\alpha_{\rm NR}(\omega)}{d\omega^2}\right|_{\omega_0}(\omega-\omega_0)^2 + \Delta\alpha(\omega_0) + \left.\frac{d\Delta\alpha(\omega)}{d\omega}\right|_{\omega_0}(\omega-\omega_0) = 0 \quad (2)$$

   for the unknown first-order correction $\Delta\omega_1 = \omega - \omega_0$.

3. Repeat with $\omega_1 = \omega_0 + \Delta\omega_1$ in place of ω_0 in Eq. (2), and continue to convergence. Of course, $\alpha_{\rm NR}(\omega_0) = 0$ by definition, but $\alpha_{\rm NR}(\omega_i) \neq 0$ for $i = 1, 2, 3, \cdots$.

The results are as shown in Table 1. The last column shows that the change in $\omega_{\rm TO}$ after the first iteration is only 0.5 MHz, and so Eq. (2) is sufficiently accurate as it stands without further iterations when both the first- and second-derivatives are kept. The second, third and fourth blocks of numbers in the table show what happens if the derivative of the tensor part $-\frac{1}{2}\alpha_{\rm T}$ and/or the second derivative $d^2\alpha_{\rm NR}/d\omega^2$ are set equal to zero. The convergence is slower, but the final result is unchanged. The biggest effect comes from the second derivative term. If this term is omitted, then the first iteration yields a correction of −176 MHz. This may be significant in the analysis of the experimental results.

Note that the final converged value of 725 743 950.1 MHz has changed by −28 MHz from the value 725 743 978 MHz reported previously, and so Table S1 in the SOMS should be updated accordingly. The change is due to an improved way of including the second-order finite nuclear mass effects of order $(\mu/M)^2$ and $\alpha^2\mu/M$, where μ is the electron reduced mass and M is the nuclear mass.

I have not yet calculated the vector polarizability. That will take a bit longer.

Finally, concerning my comment #16 from June 6, 2021, the situation is not as bad as stated there. I had a 500 MHz shift on interation because my second derivative term was in fact incorrect (I forgot to zero the accumulator before summing over states). However, the iterations still converge to the correct answer—it is just slower. If the first and second derivatives are correctly calculated, then the shift is only 0.5 MHz, as shown in Table 1.

Gordon Drake.

Table 1: Convergence table for the tuneout frequency for the case $\alpha(\omega) = \alpha_{\rm S}(\omega) - \frac{1}{2}\alpha_{\rm T}(\omega)$, relative to the nonrelativistic value at zero frequency $\alpha_{\rm NR}(0) = 315.820\ a_0^3$, where a_0 is the Bohr radius. The second line in each pair of lines gives the contribution to the total in the first line from the tensor part $-\frac{1}{2}\alpha_{\rm T}(\omega)$. Frequencies are in MHz.

Iteration	$\alpha(\omega)$	$d\alpha/d\omega$	$\frac{1}{2}d^2\alpha_{\rm NR}/d\omega^2$	$\omega_{\rm TO}$	$\Delta\omega_{\rm TO}$
1	–0.1069795D+00	0.1080483D–05	0.1952134D–13	725 743 949.6	0.5
	–0.1838501D–02	–0.3056378D–10			
2	–0.5049313D–06	0.1084336D–05	0.1965308D–13	725 743 950.1	
	–0.1841530D–02	–0.3073435D–10			
3	–0.2082626D–16	0.1084336D–05	0.1965308D–13	725 743 950.1	
	–0.1841530D–02	–0.3073435D–10			
4	–0.9659930D–32	0.1084336D–05	0.1965308D–13	725 743 950.1	
	–0.1841530D–02	–0.3073435D–10			
	Convergence for the case $d\alpha_{\rm T}/d\omega = 0$ and $d^2\alpha_{\rm NR}/d\omega^2 = 0$				
1	–0.1069795D+00	0.1080513D–05	0.0000000D+00	725 744 123.3	–173.2
	–0.1838501D–02	0.0000000D+00			
2	0.1878275D–03	0.1084373D–05	0.0000000D+00	725 743 950.1	
	–0.1841535D–02	0.0000000D+00			
3	0.5910408D–08	0.1084367D–05	0.0000000D+00	725 743 950.1	
	–0.1841530D–02	0.0000000D+00			
4	0.1675201D–12	0.1084367D–05	0.0000000D+00	725 743 950.1	
	–0.1841530D–02	0.0000000D+00			
	Convergence for the case $d^2\alpha_{\rm NR}/d\omega^2 = 0$				
1	–0.1069795D+00	0.1080483D–05	0.0000000D+00	725 744 126.1	–176.0
	–0.1838501D–02	–0.3056378D–10			
2	0.1908644D–03	0.1084343D–05	0.0000000D+00	725 743 950.1	
	–0.1841535D–02	–0.3073465D–10			
3	0.6059309D–09	0.1084336D–05	0.0000000D+00	725 743 950.1	
	–0.1841530D–02	–0.3073435D–10			
4	0.6106912D–20	0.1084336D–05	0.0000000D+00	725 743 950.1	
	–0.1841530D–02	–0.3073435D–10			
	Convergence for the case $d\alpha_{\rm T}/d\omega = 0$				
1	–0.1069795D+00	0.1080513D–05	0.1952134D–13	725 743 946.8	3.3
	–0.1838501D–02	0.0000000D+00			
2	–0.3525580D–05	0.1084366D–05	0.1965307D–13	725 743 950.1	
	–0.1841530D–02	0.0000000D+00			
3	–0.9992699D–10	0.1084367D–05	0.1965308D–13	725 743 950.1	
	–0.1841530D–02	0.0000000D+00			
4	–0.2832244D–14	0.1084367D–05	0.1965308D–13	725 743 950.1	
	–0.1841530D–02	0.0000000D+00			