Tune-out wavelengths for meta-stable helium in linearly and circularly polarized light

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A. Tune-out wavelengths for three different polarizations for magnetic sub-level

The dynamic dipole polarizabilities for the magnetic sub-level of He ($1s2s\,^3S_{1,M_J}$) is

$$\alpha_1(\omega) = \alpha_1^0(\omega) + A\cos\theta_k \frac{M_J}{J}\alpha_1^1(\omega) + \left\{\frac{3\cos^2\theta_p - 1}{2}\right\}\frac{3M_J^2 - J(J+1)}{J(2J-1)}\alpha_1^2(\omega) \tag{1}$$

where A, θ_k and θ_p define the degree of circular polarization, the angle between the wave vector of the electric field and the z axis, and the angle between the direction of polarization and the z axis, respectively. Here $A = 0$ is the π polarization (linearly polarization), $A = 1$ means σ^+ polarization, and $A = -1$ is the σ^- polarization. The $\alpha_1^0(\omega)$, $\alpha_1^1(\omega)$, and $\alpha_1^2(\omega)$, are known as the scalar, vector and tensor dipole polarizabilities, respectively. In the weak magnetic field, we can choose $\cos\theta_k = \cos\theta_p = 1$. For the metastable state of helium $1s2s\,^3S_{1,M_J}$, $J = 1$.

According to the Eq.(1), for the $A = 1$ and $M_J = 1$, the dynamic dipole polarizabilities is

$$\alpha_1(\omega) = \alpha_1^0(\omega) + \alpha_1^1(\omega) + \alpha_1^2(\omega) \tag{2}$$

which equals to the polarizabilities of $A = -1$ and $M_J = -1$. That is to say the dynamic dipole polarizabilities in different polarizations of the magnetic sub-level can equal each other. Such as

$$\alpha_1(\omega; A = 1, M_J = 1) = \alpha_1(\omega; A = -1, M_J = -1) \tag{3}$$

$$\alpha_1(\omega; A = 1, M_J = -1) = \alpha_1(\omega; A = -1, M_J = 1) \tag{4}$$

$$\alpha_1(\omega; A = 0, M_J = 1) = \alpha_1(\omega; A = 0, M_J = -1) \tag{5}$$

$$\alpha_1(\omega; A = 1, M_J = 0) = \alpha_1(\omega; A = -1, M_J = 0) = \alpha_1(\omega; A = 0, M_J = 0) \tag{6}$$

So in order to simplify our calculations, we will just calculate the dynamic dipole polarizabilities for the following four types.

$$\alpha_1(\omega; A = 1, M_J = 1) \tag{7}$$

for the σ^+ polarization of $1s2s\,^3S_{1,1}$ sub-level,

$$\alpha_1(\omega; A = 1, M_J = -1) \tag{8}$$

TABLE I: The tune-out wavelengths for the magnetic sub-levels $1s2s\,^3S_{1,M_J}$ of helium. For each state, the first line is the tune-out wavelength in atomic units, and the second line is the tune-out wavelength in nanometer.

Sub-level	A=1	A=-1	A=0
$1s2s\,^3S_{1,1}$	0.110317779	0.110316985	0.110317382
	413.0191248	413.0220974	413.0206111
$1s2s\,^3S_{1,-1}$	0.110316985	0.110317779	0.110317382
	413.0220974	413.0191248	413.0206111
$1s2s\,^3S_{1,0}$	0.110318348	0.110318348	0.110318348
	413.0169945	413.0169945	413.0169945

for the σ^+ polarization of $1s2s\,^3S_{1,-1}$ sub-level,

$$\alpha_1(\omega; A = 0, M_J = 1) \tag{9}$$

for the π polarization of $1s2s\,^3S_{1,1}$ sub-level, and

$$\alpha_1(\omega; A = 1, M_J = 0) \tag{10}$$

for the σ^+ polarization of $1s2s\,^3S_{1,0}$ sub-level.

Once the dynamic dipole polarizabilities are obtained, then we can determine the tune-out wavelengths for the magnetic sub-levels. All the results are listed in the Table I.

B. Zeeman Shift for the magnetic sub-level of $2\,^3S_{1,M_J}$

Using the LS coupling and standard angular momentum theory, the expectation value of the Zeeman Hamiltonian is

$$\langle LSJ'M_J|H_{Zeeman}|LSJM_J\rangle = (\mu_B H)(-1)^{1-M_J}\sqrt{6(2J+1)(2J'+1)}\begin{pmatrix} J' & 1 & J \\ -M_J & 0 & M_J \end{pmatrix}$$
$$\left[(-)^{J+J'+L+S}g'_L\begin{Bmatrix} L & J' & S \\ J & L & 1 \end{Bmatrix} + (-)^{L+S}g'_S\begin{Bmatrix} J & J' & 1 \\ S & S & L \end{Bmatrix} + (-)^{J'}g_x\begin{Bmatrix} L & L & 2 \\ S & S & 1 \\ J' & J & 1 \end{Bmatrix}\right]$$
$$+\frac{2}{3}(\mu_B H)^2(-1)^{J+J'+L+S-M_J}\sqrt{(2J+1)(2J'+1)}$$
$$\left[g_{Q_1}\begin{pmatrix} J' & 0 & J \\ -M_J & 0 & M_J \end{pmatrix}\begin{Bmatrix} L & J' & S \\ J & L & 0 \end{Bmatrix} - g_{Q_2}\begin{pmatrix} J' & 2 & J \\ -M_J & 0 & M_J \end{pmatrix}\begin{Bmatrix} L & J' & S \\ J & L & 2 \end{Bmatrix}\right] \tag{11}$$

where H is the external magnetic field, and μ_B is the Bohr magneton. g'_L, g'_S, g_x, g_{Q_1}, and g_{Q_2} are five g factors, which characterize the Zeeman effect to order α^2, can be further expressed in terms of 11 reduced matrix elements according

$$g'_L = \sqrt{\frac{L(L+1)(2L+1)}{6}}g_L + \frac{2}{\sqrt{6}}\frac{m}{M}F_1 + \alpha^2\frac{1}{\sqrt{6}}(F_2+F_3-F_4) \tag{12}$$

$$g'_S = \sqrt{\frac{S(S+1)(2S+1)}{6}}g_S + \alpha^2(-1)^S\frac{(2S+1)}{\sqrt{2L+1}}\begin{Bmatrix} \frac{1}{2} & S & \frac{1}{2} \\ S & \frac{1}{2} & 1 \end{Bmatrix}(F_5 + \frac{Z}{3}F_6 - \frac{1}{2}F_7) \tag{13}$$

$$g_x = \alpha^2(-1)^S(2S+1)\sqrt{\frac{5}{6}}\begin{Bmatrix} \frac{1}{2} & S & \frac{1}{2} \\ S & \frac{1}{2} & 1 \end{Bmatrix}(-ZF_8 + \frac{3}{2}F_9) \tag{14}$$

$$g_{Q_1} = F_{10} \tag{15}$$

$$g_{Q_2} = F_{11} \tag{16}$$

where

$$g_L = 1 - m/M \tag{17}$$

$$g_S = 2[1 + \alpha/2\pi - 0.328478965(\alpha^2/\pi^2) + \cdots] \tag{18}$$

TABLE II: The Zeeman shift (MHz) for the magnetic sub-levels $1s2s\,^3S_{1,M_J}$ of helium. The magnetic field is in Gauss units.

Magnetic Field	$1s2s\,^3S_{1,1}$	$1s2s\,^3S_{1,-1}$	$1s2s\,^3S_{1,0}$
0.3344	0.468034975344753	−0.468034974835478	0.000000000254638
0.38	0.531857926567587	−0.531857925909948	0.000000000328819
2.96	4.142893340127071	−4.142893300224252	0.000000019951410
3.01	4.212874646890846	−4.212874605628573	0.000000020631137
3.06	4.282855953666007	−4.282855911021509	0.000000021322249

m/M is the electron to nuclear mass ratio, and Z is the atomic number. According to the results of He($2\,^3S_1$) from the calculations of Yan (PRA 50, R1980 (1994)), there are just g_S and g_{Q_1} non-zero, so the Zeeman shift for the magnetic sub-level of He($2\,^3S_1$) with $L=0$, $S=1$, $J'=J=1$, is written as

$$\begin{aligned}\langle LSJ'M_J|H_{Zeeman}|LSJM_J\rangle &= (\mu_B H)(-1)^{M_J}3\sqrt{6}g'_S\begin{pmatrix}1 & 1 & 1\\ -M_J & 0 & M_J\end{pmatrix}\begin{Bmatrix}1 & 1 & 1\\ 1 & 1 & 0\end{Bmatrix}\\ &+2(\mu_B H)^2(-1)^{1-M_J}g_{Q_1}\begin{pmatrix}1 & 0 & 1\\ -M_J & 0 & M_J\end{pmatrix}\begin{Bmatrix}0 & 1 & 1\\ 1 & 0 & 0\end{Bmatrix}\end{aligned}$$

$$= \sqrt{6}(-1)^{1-M_J}g'_S\begin{pmatrix}1 & 1 & 1\\ -M_J & 0 & M_J\end{pmatrix}(\mu_B H)+\frac{2\sqrt{3}}{3}(-1)^{1-M_J}g_{Q_1}\begin{pmatrix}1 & 0 & 1\\ -M_J & 0 & M_J\end{pmatrix}(\mu_B H)^2 \tag{19}$$

Adopt the value $g_S = 0.999780950608697326$ and $g_{Q_1} = 11.467588230724521$ of Yan (PRA 50, R1980 (1994)), we get the Zeeman shift in the Table II for the magnetic sub-level $2\,^3S$,

C. Zeeman Shift effect on the tune-out wavelength

According to the the laser light strength $F \simeq 70$ W/cm^2 $= 2.296567958566443\times10^4$ V/m $= 4.46611381638255\times10^{-8}$ a.u., which was centred on the trap. According to the Zeeman shift listed in the Table II, using the weighted average, the energy level of $2\,^3S_1$ state caused by magnetic field is $\Delta E_{Zeeman} = 2.546376\times10^{-4}$ Hz $= 3.87005863608\times10^{-20}$ a.u., then the total energy shift is

$$\Delta E = \Delta E_{Stark} + \Delta E_{Zeeman} = -\frac{1}{2}\alpha_1(\omega)F^2 + \Delta E_{Zeeman} \tag{20}$$

At this situation, the new tune-out wavelength ω_{to} is obtained using the followed formula,

$$\Delta E = -\frac{1}{2}\alpha_1(\omega_{to})F^2 + \Delta E_{Zeeman} = 0 \tag{21}$$

which can be simplified as

$$\alpha_1(\omega_{to}) = \frac{2\Delta E_{Zeeman}}{F^2} = 3.88050250004169 \times 10^{-5} \tag{22}$$

Using the data of matrix elements and energies in our previous paper (Mitroy and Tang), we get the new tune-out wavelength

$$\omega_0 = \lambda_{to} = 0.1103173876655669\ a.u. = 413.020589880494965393478631904060\ nm \tag{23}$$

compared with value previous theoretical value 413.019405 nm , we can see the Zeeman shift effect on the tune-out wavelength is $(413.02058988049496539347863190406 - 413.019405) = 0.0011848804949\ nm \simeq 1.2\ pm$. In other word, the effect on the tune-out wavelength from the Zeeman shift is about 3 ppm.

D. Derivation of the dynamic dipole polarizabilities as the frequency

Since the dynamic dipole polarizabilities is

$$\alpha_1(\omega) = \sum_n \frac{f_{0n}}{\Delta E_{0n}^2 - \omega^2} \tag{24}$$

So the first-order derivation of dynamic dipole polarizabilities is

$$\begin{aligned}
\frac{d\alpha_1(\omega)}{d\omega} &= \sum_n f_{0n}\left[\frac{1}{\Delta E_{0n}^2 - \omega^2}\right]' \\
&= \sum_n f_{0n}\left[(\Delta E_{0n}^2 - \omega^2)^{-1}\right]' \\
&= \sum_n f_{0n}\left[-(\Delta E_{0n}^2 - \omega^2)^{-2} \times (-2\omega)\right] \\
&= \sum_n \frac{2\omega f_{0n}}{(\Delta E_{0n}^2 - \omega^2)^2}
\end{aligned} \tag{25}$$

which presents the tangential line or the slope of the dipole polarizabilities.

The second-order derivation of dynamic dipole polarizabilities is

$$
\begin{aligned}
\frac{d^2\alpha_1(\omega)}{d\omega^2} &= \frac{d}{d\omega}\left[\frac{d\alpha_1(\omega)}{d\omega}\right] \\
&= \frac{d}{d\omega}\left[\sum_n \frac{2\omega f_{0n}}{(\Delta E_{0n}^2-\omega^2)^2}\right] \\
&= \sum_n 2f_{0n}\frac{d}{d\omega}\left[\frac{\omega}{(\Delta E_{0n}^2-\omega^2)^2}\right] \\
&= \sum_n 2f_{0n}\left\{\frac{1}{(\Delta E_{0n}^2-\omega^2)^2}+\omega\frac{d}{d\omega}\left[(\Delta E_{0n}^2-\omega^2)^{-2}\right]\right\} \\
&= \sum_n 2f_{0n}\left[\frac{1}{(\Delta E_{0n}^2-\omega^2)^2}+4\omega^2(\Delta E_{0n}^2-\omega^2)^{-3}\right] \\
&= \sum_n \frac{2f_{0n}}{(\Delta E_{0n}^2-\omega^2)^2}+\sum_n \frac{8\omega^2 f_{0n}}{(\Delta E_{0n}^2-\omega^2)^3}
\end{aligned}
\tag{26}
$$

which presents the concavity of the dynamic dipole polarizabilities. When this value is positive, then the polarizabilities has minimum, otherwise, the polarizabilities has maximum.

The third-order derivation of dynamic dipole polarizabilities is

$$
\begin{aligned}
\frac{d^3\alpha_1(\omega)}{d\omega^3} &= \frac{d}{d\omega}\left\{\sum_n \frac{2f_{0n}}{(\Delta E_{0n}^2-\omega^2)^2}+\sum_n \frac{8\omega^2 f_{0n}}{(\Delta E_{0n}^2-\omega^2)^3}\right\} \\
&= \sum_n 2f_{0n}\frac{d}{d\omega}\left[(\Delta E_{0n}^2-\omega^2)^{-2}\right]+\sum_n 8f_{0n}\frac{d}{d\omega}\left[\frac{\omega^2}{(\Delta E_{0n}^2-\omega^2)^3}\right] \\
&= \sum_n 2f_{0n}\left[\frac{4\omega}{(\Delta E_{0n}^2-\omega^2)^3}\right]+\sum_n 8f_{0n}\left[\frac{2\omega}{(\Delta E_{0n}^2-\omega^2)^3}+\frac{6\omega^3}{(\Delta E_{0n}^2-\omega^2)^4}\right] \\
&= \sum_n \frac{24\omega f_{0n}}{(\Delta E_{0n}^2-\omega^2)^3}+\sum_n \frac{48\omega^3 f_{0n}}{(\Delta E_{0n}^2-\omega^2)^4}
\end{aligned}
\tag{27}
$$

The fourth-order derivation of dynamic dipole polarizabilities is

$$\begin{aligned}
\frac{d^4\alpha_1(\omega)}{d\omega^4} &= \frac{d}{d\omega}\left[\sum_n \frac{24\omega f_{0n}}{(\Delta E_{0n}^2-\omega^2)^3}+\sum_n \frac{48\omega^3 f_{0n}}{(\Delta E_{0n}^2-\omega^2)^4}\right] \\
&= \sum_n 24 f_{0n}\frac{d}{d\omega}\left[\frac{\omega}{(\Delta E_{0n}^2-\omega^2)^3}\right]+\sum_n 48 f_{0n}\frac{d}{d\omega}\left[\frac{\omega^3}{(\Delta E_{0n}^2-\omega^2)^4}\right] \\
&= \sum_n 24 f_{0n}\left[\frac{1}{(\Delta E_{0n}^2-\omega^2)^3}+\frac{6\omega^2}{(\Delta E_{0n}^2-\omega^2)^4}\right]+\sum_n 48 f_{0n}\left[\frac{3\omega^2}{(\Delta E_{0n}^2-\omega^2)^4}+\frac{8\omega^4}{(\Delta E_{0n}^2-\omega^2)^5}\right] \\
&= \sum_n \frac{24 f_{0n}}{(\Delta E_{0n}^2-\omega^2)^3}+\sum_n \frac{288\omega^2 f_{0n}}{(\Delta E_{0n}^2-\omega^2)^4}+\frac{384\omega^4 f_{0n}}{(\Delta E_{0n}^2-\omega^2)^5}
\end{aligned} \tag{28}$$

According to the data of dynamic dipole polarizabilities of metastable helium, we obtained the derivatives of the polarizability (the units is a_0^3/nm) with wavelength around the tune out is

$$\begin{aligned}
\frac{d\alpha(\omega)}{d\lambda} &= -1.913 \\
\frac{d^2\alpha(\omega)}{d\lambda^2} &= -454.79 \\
\frac{d^3\alpha(\omega)}{d\lambda^3} &= -2.04\times 10^5 \\
\frac{d^4\alpha(\omega)}{d\lambda^4} &= -1.19\times 10^8
\end{aligned} \tag{29}$$

E. Determination of the finite nuclear mass effect and relativistic correction on the tune-out wavelength

Here we calculate the tune-out wavelength for metastable $^\infty$He, ^{4}He, and ^{3}He isotopes by using the nonrelativistic Hylleraas variational method. Also the relativistic effect on the static polarizabilities and tune-out wavelengths are calculated using the B-spline relativistic CI method (at moment the value is smallest basis sets, new values for the bigger basis sets are calculating, I will update it once the calculation finished) . All the results combined our theoretical values published in 2003 are listed in the Table III. From the last three lines, we can see the finite nuclear mass effect on the static dipole polarizability from the first decimal digits, and the finite nuclear mass effect about 0.1-0.15 nm on the tune-out wavelength. It seems that the relativistic effect decreased the value of $\alpha_1(0)$ and the tune-out

TABLE III: Comparison of the static dipole polarizability and the tune-out wavelengths of $2\,^3S$. The last three lines are the results from Hylleraas variational method. The third- and forth-column are the tune-out wavelength in atomic units and in the unit of nanometer.

Methods	$\alpha_1(0)$	a.u.	nm
RCI $^\infty He$	315.54276 714	0.110 325 4633	412.990 3575
Hybrid $^\infty He$	315.462	0.110 317 7042	413.019 4050
CPM $^\infty He$	316.020	0.110 375 8821	412.801 707
$^\infty$He	315.63147(1)	0.110 312 6(2)	413.0383(2)
^{4}He	315.82041(1)	0.110 285 6(2)	413.1392(11)
^{3}He	315.88223(1)	0.110 276 9(3)	413.1723(12)

wavelength, but the finite nuclear mass effect increased them. In fact, the relativistic effect is not cancelled by the finite nuclear mass correction, this point is different with the static dipole polarizabilities of ground-state of helium (for ground-state, two terms are cancelled each other). So the experimental result 413.0878(9) lies within the values of RCI and ^{3}He.

F. The effect of broadband light on the tune-out wavelength

According to the paper of Cronin et al (PRL 109, 243004 (2012)), there is a paragraph (see followed), which stated how they considered the broadband light effect on the tune-out wavelength,

After spatial filtering with a single mode fiber, 1% of the power was in a broadband spectral component from spontaneous emission in the tapered amplifier. To quantify the uncertainty in λ_{zero} caused by this broadband component, we characterized the laser spectrum with a grating spectrometer, and we accounted for the laser spectrum by modifying the Eq.(30) as Eq.(31) with an additional integral over the frequency-dependent laser intensity. We calculated that the broadband light introduces an uncertainty of 0.5 pm to our measurement of λ_{zero} zero.

$$\phi_0(\omega) = \frac{\alpha(\omega)}{2\epsilon_0 c\hbar\upsilon}\int_{-\infty}^{\infty} I(x,z)dz = b\alpha(\omega) \tag{30}$$

$$\phi(\omega) = \frac{1}{\epsilon_0 c\hbar v}\int_{z=-\infty}^{\infty}\int_{\omega=0}^{\infty}\alpha_(\omega)I(\omega;x,z)dzd\omega \tag{31}$$

So how to calculate the effect from broadband light? I think the key point is we should know which frequency-dependent physical quantities are closely related to our measurement. In Cronin's experiment, they get the tune-out wavelength of the K atom by measuring the phase shift $\phi(\omega)$, which not only depend on the dynamic dipole polarizability $\alpha(\omega)$, but also depend on the laser beam intensity $I(\omega;x,z)$. So in the Fig 5.8 of Holmgren's 2013 thesis, he gave the curves of the dynamic dipole polarizability and the power as the frequency ω, inserting the values of the curves into Eq.(31), he obtained a new cure of phase shift to extract a new tune-out wavelength, compared with their previous result by considering an ideal monochromatic laser spectroscopy, then they got the effect of broadband light on the tune-out wavelength.

Here since I didn't have the data file of $I(\omega;x,z)$ of Cronin et al, so I can't repeat their calculations.

Next, I try to recall how Bryce deal with this problem. The first order in polarizability the signal seen by our experiment is proportional to

$$Signal(\lambda) = k\int_{-\infty}^{\infty} I(\lambda)(\lambda-\lambda_0)d\lambda \tag{32}$$

where $I(\lambda)$ is the laser spectrum, which obtained by using three different deconvolution algorithms, λ_0 is the tune-out wavelength, and k is the proportion factor. Bryce assumed that the laser spectrum will translated at different laser wavelengths, so used the laser wavelength that was used to take these spectra 413.08784nm for λ_0, and add in a shift factor to the laser spectrum,

$$Signal(\lambda_{shift}) = \int_{-\infty}^{\infty} I(\lambda)(\lambda-\lambda_{shift})(\lambda-\lambda_{center})d\lambda \tag{33}$$

Then find the x intercept of $Signal(\lambda_{shift})$ to get the λ_{shift}, which corresponds to the wavelength shift in the tune-out due to the spectral distribution. His results from three different deconvolution algorithm are followed

$$RichardsonLucy = -0.00481688\,nm \tag{34}$$

$$Wiener = -0.00336092\,nm \tag{35}$$

$$SteepestDescent = 0.00198908\,nm \tag{36}$$

So my questions about Bryce's method are

1. The potential depth felt by atom is in proportion to the polarizability not to the wavelength. So in the above Eq.(32), how to understand your assumption: "the first order in polarizability the signal seen by our experiment is proportional to $(\lambda - \lambda_0)$ although it's clearly seen from the Fig.4 of the manuscript you sent to me the last time?

2. In the Eq.(33), why we need add the factor $(\lambda - \lambda_{center})$ and what's the value of λ_{center} you adopted?

In my opinion, since you have all the data of Fig.4 of the manuscript, which is the curve of the amplitude as the laser wavelength, also you have the data file of the laser spectrum from three different deconvolution algorithms, you can multiply two data file then give a new curve as the wavelength, then extract a new tune-out wavelength, comparing with the 413.0878(9) nm, the effect from broadband spectrum can be obtained. This method is the same as the Cronin's.