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Collisional Effects on the Dipole Transitions of Pionic Helium*

B. Obreshkov
Institute for Nuclear Research and Nuclear Energy,
Bulgarian Academy of Sciences, Sofia 1784, Bulgaria

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Abstract. The collision-induced shift and broadening of selected dipole transition lines of pionic helium in gaseous helium at low temperatures up to $T = 12$ K and pressure up to a few bar are calculated in the approximation of binary collisions. We predict blue shifts of the resonance frequencies of the unfavored and red shifts for the favored transitions. The collisional Stark mixing and quenching of the metastable states of the pionic atom is found to be unlikely at thermal energies due to the large separation between the energy levels in the pionic atom within the same energy shell.

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1 Introduction

Pionic helium is a three-body system composed of a helium nucleus, an electron in a ground state and π^- in highly excited nearly circular state with principal and orbital quantum numbers $n \sim l + 1$. These states can de-excite via Auger transitions to lower lying states which have large overlap with the helium nucleus and subsequently undergo fast nuclear absorption for times less than 10^{-12} s. However long-lived π^- were observed in bubble-chamber experiments [1]. To explain this anomaly, Condo suggested that metastable atomic states of π^- are formed

$$\mathrm{He} + \pi^- \to [\mathrm{He}^+\pi^-]_{nl} + e^- \tag{1}$$

in which π^- occupies highly excited Rydberg states with principal quantum number $n \sim (m^*/m_e)^{1/2}$, where m^* is the reduced mass of π^- and the helium nucleus. Thus, $n \sim 16$ for initially occupied states by π^-. For nearly circular orbits $n \sim l + 1$, the Auger decay rate is strongly suppressed and because the radiative decay $\tau_{\mathrm{rad}} \sim 1\ \mu s$ is also slow, the lifetime of the metastable states is determined by the proper lifetime of π^-, $\tau_{\pi^-} \sim 26$ ns. An indirect conformation

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of the Condo's hypothesis has been obtained at TRIUMF [2] in experiments with π^- stopped in liquid helium; it has been found that about 2% of the pions retain a lifetime of 7 ns.

When comparing experimental transition frequencies to three-body QED calculations of pionic helium, the π^- mass can be determined with fractional precision better than 1 ppm [3]. Systematic effects such as collision-induced shift and broadening of the transition lines, the collision quenching of the metastable pionic states, AC Stark shifts, frequency chirp in the laser beam, can prevent the experiment from achieving this high precision.

At higher helium target densities, the elastic scattering of $\pi^-\mathrm{He}$ atoms with atoms of the helium gas cause shift and broadening of transition wavelengths. The Stark mixing of the metastable pionic states with states that are short lived due to Auger process, leading to a subsequent fast nuclear absorption of π^-. If the collisional de-excitation rates become comparable to the radiative ones, the lifetime of some of the metastable states might be shortened. Reliable theoretical calculation for the density-dependent shift and width of the dipole transition line shapes and the quenching rates of the metastable pionic atom states is needed for the interpretation of the experimental data and extrapolation of transition wavelengths at zero density of the helium gas. In the present paper we present numerical results for the collisional shift and broadening of the dipole transition lines in pionic helium due to elastic scattering with ordinary helium gas atoms, and evaluate perutrbatively the quenching cross sections for the transitions $(n,l) \rightarrow (n,l-1)$ due to collisional Stark mixing involving pionic atom states within the same energy shell n

$$[\mathrm{He}^+\pi^-]_{nl} + \mathrm{He} + \varepsilon \rightarrow [\mathrm{He}^+\pi^-]_{nl-1} + \mathrm{He} + \varepsilon' \tag{2}$$

where ε and ε' are the initial and final kinetic energies of the relative motion of the colliding atoms.

2 Collisional Shift and Broadening Calculations

2.1 Interatomic potentials

The collisional shift and broadening of the laser stimulated transition line in pionic helium are obtained in the impact approximation of the binary collision theory of the spectral line shape [5–8].

This approach was already applied in the calculations of the density effects on the line shape in antiprotonic helium [9–11] and produced theoretical results in agreement with experiment [12], see also [13] and references therein. The success of these calculations was due to the use of a highly accurate three-electron potential energy surface (PES) for the description of the binary interaction of an exotic helium atom with the atoms of the helium gas. The PES had been evaluated with *ab initio* quantum chemistry methods [14] for nearly 400 configurations of the three heavy constituents of the interacting atoms (two helium nuclei and an antiproton or pion), selected to match the typical interparticle distances in the metastable states of exotic helium of experimental interest [15]. The nuclear configurations are parameterized with the length r of the vector joining the heavy particles in the pionic atom, the length R of the vector joining its center-of-mass with the nucleus of the perturbing helium atom, and the angle θ between them. Subsequently, the numerical values of the PES at these 400 grid points were fitted with smooth functions $V(r, R, \theta)$ and used in the calculation of the collisional shift and dephasing rate. Because the collisional quenching due to inelastic collisions at thermal energies is unlikely to be relevant for the metastable states of the pionic atom, we use central state-dependent potentials to describe the elastic scattering

$$V_{nl}(R) = \frac{1}{2} \int dr |\chi_{nl}(r)|^2 \int d\theta \sin\theta V(r, R, \theta), \tag{3}$$

where $\chi_{nl}(r)$ are the unperturbed π^- wave-functions. The effect of the detailed angular-dependent part of the interaction energy has shown to lead to minor corrections to the line shift and width.

2.2 Impact approximation

When the helium gas atoms are moving rapidly, the broadening and shift of the spectral lines arises from a series of binary encounters between the pionic atom with ordinary helium atom. The impact approximation is valid if the average time interval between collisions is much larger than the duration of the collision [6]. Table 1 shows the state-dependent elastic scattering cross-section and collision rates at temperature $T = 6$ K and number-density $N = 10^{21}$ cm^{-3} of the perturbing helium gas atoms. The time interval between the collisions is of order of $\tau_c \approx 10$ picoseconds, that is an order a magnitude shorter than the duration of the collision $\tau_d < 1$ ps, which validates the applicability of the impact approximation at thermal collision energies.

Table 1. Thermally averaged elastic scattering cross sections σ (in units 10^{-15} cm^2) and rates $\lambda = N\langle v\sigma\rangle$ (in units s^{-1}) at temperature 6K and number density of the helium gas 10^{21} cm^{-3}. The notation $a[b] = a \times 10^b$ is used.

(n, l)	σ_{nl}	λ_{nl}
$16, 14$	2.9	0.72 [11]
$16, 15$	0.8	0.78 [11]
$17, 16$	3.5	0.87 [11]
$17, 15$	4.4	1.10 [11]

For an isolated spectral line produced by a laser-stimulated dipole transition from an initial state $i = (n, l)$ to a final state $f = (n', l')$ of the pionic atom, the line shape assumes a Lorentizan profile

$$I_{fi}(\omega) = \frac{|_{fi}|^2}{\pi} \frac{\Gamma_{fi}}{(\omega - \omega_{fi} + \Delta_{fi})^2 + \Gamma_{fi}^2}, \tag{4}$$

where $_{fi}$ is the transition dipole moment, $\omega_{fi} = E_f - E_i$ is the transition frequency and Γ_{fi} and Δ_{fi} are the collision-induced broadening and shift, respectively. In the approximation of binary collisions, both the shift $\Delta_{fi} = N\beta_{fi}$ and the polarization dephasing rate $\Gamma_{fi} = N\alpha_{fi}$ are linear functions of the gas density. The slope of the temperature-dependent collisional broadening and shift are

$$\alpha_{fi}(T) = \left\langle \frac{\pi}{Mk} \sum_{L=0}^{\infty} (2L+1) 2 \sin^2 \eta_{fi,L}(k) \right\rangle_T \tag{5}$$

and

$$\beta_{fi}(T) = -\left\langle \frac{\pi}{Mk} \sum_{L=0}^{\infty} (2L+1) \sin 2\eta_{fi,L}(k) \right\rangle_T, \tag{6}$$

respectively, where $k = Mv$ is the wave-number of relative motion, v is the impact velocity and M is the reduced mass of the collision system. Both α and β and expressed in terms of elastic scattering phase shifts $\eta_{fi,L}(k) = \delta_{iL}(k) - \delta_{fL}(k)$ and $\langle F \rangle_T$ is a thermal average

$$\langle F \rangle_T = 4\pi \left(\frac{M}{2k_B T} \right)^{3/2} \int dv v^2 e^{-Mv^2/2k_B T} F(v) \tag{7}$$

over the Maxwell velocity distribution at temperature T for the pionic helium-helium system and k_B is the Boltzmann constant. The partial wave phases $\delta_L(k) = \delta_L(k, R \to \infty)$ are obtained from the asymptotic solution of the variable phase equation [17]

$$\begin{aligned} \frac{d}{dR} \delta_{nlL}(k, R) = -\frac{2MV_{nl}(R)}{k} \Big[& \cos \delta_{nlL}(k, R) j_L(kR) \\ & - \sin \delta_{nlL}(k, R) n_L(kR) \Big]^2, \end{aligned} \tag{8}$$

subject to the boundary condition $\delta_{nlL}(k, 0) = 0$, and $j_L(z), n_L(z)$ are the Riccati-Bessel functions.

Tables 2-5 present values for the scattering phase shifts $\eta_{fi,L}(k)$ for the "unfavored" transitions $(16, 15) \to (16, 14)$, $(16, 15) \to (17, 14)$ and $(17, 16) \to (17, 15)$, and for the "favored" one $(17, 16) \to (16, 15)$. respectively. For the unfavored transition $(16, 15) \to (16, 14)$ in Table 2, scattering phases are appreciably less than 1 radian over the whole range of wave numbers. In this regime of weak collisions, the phase shifts are added linearly and contribute to the line shift Δ, but have little effect on the broadening Γ. Since all phases are negative, the transition frequency undergoes a blue shift. For $k < 0.5$, the scattering of s, p and d- -waves gives dominant contribution to the dipole transition lineshape, contributions of partial waves with $L > 3$ are suppressed due to large centrifugal barrier. Larger number of partial waves is required to converge Δ and Γ for $k > 0.5$. Similar results are found for the other two unfavored transitions $(16,15) \rightarrow (17,14)$ in Table 3 and $(17,16) \rightarrow (17,15)$ in Table 4, state-dependent scattering phases are negative over the whole range of wavenumbers, but have appreciable values resulting in substantial broadening of the dipole transition lines. Table 3 shows that the s-wave phase shift $\eta_0 \rightarrow -\pi$ near $k \rightarrow 0$, because the scattering potential in the final state $V_{17,14}(R)$ supports a single bound state. Because the s-wave scattering is dominant towards threshold, this bound state affects dramatically the transition line shape at very low speeds with $k \leq 0.1$ as the center frequency undergoes a red shift when $\eta_0 < -\pi/2$. However the thermally-averaged shift and width are weakly affected by the low-velocity tail in the Maxwell distribution. Tables 3 and 4 show that at thermal energies, the contribution of close-range binary encounters involving s- and p-wave scattering is enhanced, but d- and f-wave scattering phase shifts involving more distant collisions are essential due to their larger statistical weight $2L + 1$. The elastic scattering phase shifts associated with the favored transition are shown in Table 5, scattering phases are positive resulting in redshift of the line-center due to contributions of s, p and d-wave elastic scattering.

Table 6 presents numerical results on the temperature dependence of the slopes of the collisional shift and broadening, $\beta_{fi}(T)$ and $\alpha_{fi}(T)$, of the two transition lines of known experimental interest [16]. The temperature dependence of the line profile is relatively weak in gaseous helium. At low perturber density $N = 10^{21}$ cm^{-3}, the resonance frequency of the unfavored transitions $(n,l) = (16,15) \rightarrow (16,14)$, $(16,15) \rightarrow (17,14)$ and $(17,16) \rightarrow (17,15)$ are blue shifted with $\Delta \approx 2.5$ GHz, $\Delta = 18$ GHz and $\Delta = 6$ GHz, respectively. For the favored transition $(17,16) \rightarrow (16,15)$ the line center undergoes a red-

Table 2. Phase analysis of the collisional shift and broadening of dipole transition line shape $(n,l) = (16,15) \rightarrow (n',l') = (16,14)$ in pionic helium interacting with gaseous helium at temperature $T = 6$ K.. Partial phase shifts $\eta_{fi}(k), L = 0, \ldots, 8$ in radians, k is the wave-number of relative motion.

$L = k$ a.u.	0	1	2	3	4 $\eta_{fi}(k)$, rad	5	6	7	8
0.127	-0.126	-0.047	-0.001	0.000	0.000	0.000	-0.000	0.000	0.000
0.310	-0.063	-0.067	-0.041	-0.007	-0.001	0.000	0.000	0.000	0.000
0.538	-0.047	-0.048	-0.047	-0.036	-0.014	-0.004	-0.001	0.000	0.000
0.805	-0.044	-0.043	-0.041	-0.038	-0.032	-0.019	-0.009	-0.004	-0.002
1.131	-0.046	-0.045	-0.043	-0.040	-0.035	-0.030	-0.022	-0.013	-0.007

Table 3. Phase analysis of the collisional shift and broadening of dipole transition line shape $(n,l) = (16,15) \to (n',l') = (17,14)$ in pionic helium interacting with gaseous helium at temperature $T = 6$ K. Partial phase shifts $\eta_{fi}(k)$, $L = 0,\ldots,8$ in radians, k is the wave-number of relative motion.

$L = k$	0	1	2	3	4	5	6	7	8
a.u.					$\eta_{fi}(k)$, rad				
0.127	-0.833	-0.524	-0.006	0.000	0.000	0.000	0.000	0.000	0.000
0.310	-0.484	-0.508	-0.287	-0.026	-0.003	0.000	0.000	0.000	0.000
0.538	-0.407	-0.401	-0.376	-0.244	-0.058	-0.012	-0.004	-0.001	0.000
0.805	-0.401	-0.391	-0.370	-0.328	-0.234	-0.098	-0.029	-0.010	-0.005
1.131	-0.429	-0.419	-0.400	-0.369	-0.322	-0.248	-0.147	-0.062	-0.025

Table 4. Phase analysis of the collisional shift and broadening of dipole transition line shape $(n,l) = (17,16) \to (n',l') = (17,15)$ in pionic helium interacting with gaseous helium at temperature $T = 6$ K. Partial phase shifts $\eta_{fi}(k)$, $L = 0,\ldots,8$ in radians, k is the wave-number of relative motion.

$L = k$	0	1	2	3	4	5	6	7	8
a.u.					$\eta_{fi}(k)$, rad				
0.127	-0.252	-0.169	-0.002	0.000	0.000	0.000	0.000	0.000	0.000
0.310	-0.126	-0.147	-0.100	-0.002	0.000	0.000	0.000	0.000	0.000
0.538	-0.090	-0.093	-0.099	-0.081	-0.026	-0.007	-0.001	0.000	0.000
0.805	-0.077	-0.077	-0.077	-0.068	-0.038	-0.015	-0.006	-0.003	-0.001
1.131	-0.073	-0.072	-0.071	-0.068	-0.065	-0.059	-0.045	-0.026	-0.014

Table 5. Phase analysis of the collisional shift and broadening of dipole transition line shape $(n,l) = (17,16) \to (n',l') = (16,15)$ in pionic helium interacting with gaseous helium at temperature $T = 6$ K. Partial phase shifts $\eta_{fi}(k)$, $L = 0,\ldots,8$ in radians, k is the wave-number of relative motion.

$L = k$	0	1	2	3	4	5	6	7	8
a.u.					$\eta_{fi}(k)$, rad				
0.127	0.382	0.115	0.000	0.000	0.000	0.000	0.000	0.000	0.000
0.310	0.243	0.226	0.083	0.002	-0.002	0.000	0.000	0.000	0.000
0.538	0.230	0.219	0.183	0.087	0.009	0.000	0.000	0.000	0.000
0.805	0.248	0.238	0.217	0.177	0.103	0.025	0.006	-0.001	0.000
1.131	0.280	0.273	0.257	0.231	0.192	0.133	0.061	0.013	0.000

shift with $\Delta \approx -8$ GHz. The large collisional broadening of the transition line $(16,15) \to (17,14)$ $\Gamma = 7.7$ GHz would not allow the transition wavelength to be determined to a fractional precision better than 1 ppm. Because the collisional broadening of the transition line $(16,15) \to (16,14)$ is $\Gamma = 0.1$ GHz, this dipole transition is suitable for spectroscopic measurements in pionic helium.

Table 6. Slope of the density shift and broadening $\beta(\alpha)$ for selected transitions in pionic helium and temperatures in the range $4-12$ K, in units of 10^{-21} GHz.cm^3

	Temperature (K)				
Transition	4	6	8	10	12
$(16,15) \rightarrow (16,14)$	2.97(0.16)	2.93(0.14)	2.92(0.13)	2.93(0.12)	2.94(0.11)
$(16,15) \rightarrow (17,14)$	17.56(7.73)	17.97(7.19)	17.95(6.98)	17.94(6.68)	18.07(6.42)
$(17,16) \rightarrow (17,15)$	6.41(0.78)	6.00(0.68)	6.16(0.64)	6.04(0.59)	5.94(0.54)
$(17,16) \rightarrow (16,15)$	-7.45(1.55)	-7.55(1.41)	-7.78(1.43)	-8.00(1.47)	-8.23(1.52)

3 Quenching Cross-Section Calculation

The cross-sections for collisional Stark -mixing and the corresponding thermally averaged rates are given by [18]

$$\sigma_{nl,nl-1}(v) = \frac{\pi}{k^2(2l+1)} \sum_{JLL'} (2J+1)|T^J_{nlL',nl-1L}(v)|^2 \tag{9}$$

and

$$\lambda_{nl,nl-1}(T) = N\langle v\sigma_{nl,nl-1}(v)\rangle_T, \tag{10}$$

where J is the total angular momentum of the colliding atoms, $k = Mv$ and $T^J_{fi}(v)$ are the T-matrix elements. At temperatures in the range $6-300$ K the thermal collision energies ε are of the order of $10^{-5}-10^{-4}$ a.u. are very small as compared to the energy separation of the metastable states of the pionic helium (of order of 0.03 a.u. for states within the same shell n, and still larger for states with different n). Under these circumstances we evaluated the Stark-mixing amplitudes and T-matrix elements using perturbation theory.

Table 7 shows the thermally averaged cross sections and transition rates for two selected initial states of $\pi^-\mathrm{He}^+$. The inelastic cross-section of order 10^{-30} cm^2 with corresponding rates $\lambda \sim 10^{-5}$ s^{-1} makes evident that collisional quenching of the metastable states of the pionic helium is highly unlikely at thermal energies. That is because the pion excitation energy is spent to increase the kinetic energy of relative motion, which produces rapid oscillations of the final scattering state wave-function in the interaction region of the two atoms, resulting in practically vanishing transition amplitudes and quenching cross-sections.

4 Conclusion

We calculated the density shift and broadening of selected dipole transition lines in pionic helium in gaseous helium. At thermal collision energies, we find blueshift of the line center of the unfavored transitions, the transition frequency is red-shifted for the favored transition. The negligible collisional broadening

Table 7. Thermally averaged collisional quenching cross sections σ (in units 10^{-30} cm^2) and rates λ (in units s^{-1}) at temperature 6K and number density of the helium gas 10^{21} cm^{-3}. The notation $a[b] = a \times 10^b$ is used.

$(n,l) \to (n',l')$	$\sigma(nl \to n'l')$	$\lambda(nl \to n'l')$
$16,15 \to 16,14$	2.6	$5.3[-5]$
$17,16 \to 17,15$	0.8	$1.7[-5]$

($\Gamma = 0.1$ GHz) of the resonance transition $(n,l) = (16,15) \to (16,14)$ makes it suitable for precision spectroscopy of pionic helium atoms. The collisional quenching of the metastable states of pionic helium is found to be unlikely at thermal collision energies. The theoretical result may be helpful in reducing the systematic error in proposed future experiments for determination of the negatively charged pion mass from laser spectroscopy of metastable pionic helium atoms.

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