Laser induced optical activity in the region of autoionizing states

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Abstract. Optical activity of atomic medium in the region of laser coupled autoionizing states is studied theoretically. Analytical expressions and numerical results are obtained for the laser induced polarization rotation angle and the acquired ellipticity of the VUV radiation in the region of the He $2s^22p^1P$ autoionizing state (60.13 eV) coupled by circularly polarized laser field with the $2p^2S$ or $2s3d^1D$ states. A rotation angle of order $\sim 10^\circ$ can be realized for optically transparent helium media at laser intensities of $10^{10} - 10^{12}$ W/cm$^2$. Controlling the optical activity in the VUV by manipulating the laser intensity and frequency is discussed.

1. Introduction

Optical activity is the ability of a substance to change the polarization of a beam of light that is passed through it. The natural optical activity is inherent to substances with chirality and is caused by different refraction indices for the right and left circularly polarized components of the passing radiation. The optical activity can be induced naturally inactive media, for example, by magnetic and electric fields (magneto- and electro-optical effects [1, 2], or by bathing the medium in a circularly polarized laser field. In the latter case, changing the polarization properties of the probe light has been studied in the visible regime, both experimentally and theoretically, when a circularly polarized laser field couples discrete atomic states (for example [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13]). In particular, controlling the polarization rotation of the probe light by manipulating the coupling field was demonstrated [9, 12]. The highly sensitive method of optical rotation helped to obtain accurate values of optical oscillator strengths [14] and to observe for the first time [15] so-called “laser induced continuum structures” (LICS): resonances in the continuum, generated by coupling the continuum to a discrete state [16, 17, 18]. It was demonstrated with hydrogen as an example that a few overlapping LICs can lead to a more efficient control of the optical activity [19]. With the development of high-brilliant sources of VUV and soft X-ray radiation, optical activity in this wavelength range has been observed in [14, 20, 21].

We suggest to induce the optical activity in the VUV by irradiating a gas by circularly polarized IR or visible laser light coupling two autoionizing states (AIS) [22, 23]. So far the coupling of two AIS by a laser field was theoretically considered in the context of integral ionization cross sections [24, 25, 26, 27, 28] and photoelectron angular distributions [29, 30]. We
are not aware of experiments and other theoretical predictions for the laser induced polarization rotation in the VUV for such a scheme. Using as a showcase the AIS He $2s2p^1P$ coupled to another AIS, the goal of this paper is to obtain analytical expressions for the polarization rotation angle and the acquired ellipticity, to estimate the scale of the effect, to demonstrate the general features, and to control the optical activity in the VUV by manipulating frequency and intensity of the coupling laser field.

In section 2 we derive, for particular excitation schemes in He, equations for the polarization rotation angle and the acquired ellipticity. The numerical results are presented and discussed in section 3, which is followed by concluding remarks.

Unless otherwise indicated, atomic units are used throughout the paper.

2. Theory

The optical activity of a medium is determined by its susceptibilities $\chi^{(+)}$ and $\chi^{(-)}$ for the right (helicity +1) and left (helicity −1) circularly polarized components of the probe radiation, respectively. For a dilute medium ($N|\chi^{(\pm)}| \ll 1$, where $N$ is the atom number density) the initially linearly polarized probe radiation acquires the ellipticity

$$\epsilon = \pi kLN \text{Im} \left( \chi^{(-)} - \chi^{(+)} \right)$$

and its plane of polarization is rotated by the angle

$$\theta = \pi kLN \text{Re} \left( \chi^{(-)} - \chi^{(+)} \right)$$

when passing through the medium. Here $k$ is the probe field wavenumber, and $L$ is the length of the medium. The ellipticity (1) is defined as the ratio of the short and long axes of the polarization ellipse, and its sign has been chosen in such a way that $\epsilon = +1$ corresponds to right circularly polarized radiation. The positive rotation direction ($\theta > 0$) is counterclockwise when looking into the radiation source.

The probe and the laser beams are assumed to be collinear and directed along the quantization z-axis. These fields are described classically as

$$F(t) = \frac{F}{2}[e_p \exp(-i\Omega t) + \text{c.c.}], \quad f(t) = \frac{f}{2}[e_L \exp(-i\omega t) + \text{c.c.}],$$

where $F$, $\Omega$ and $e_p$ are the amplitude, the frequency, and the polarization vector of the probe field, respectively; $f$, $\omega$, and $e_L$ are those for the laser field.

The linear (with respect to the probe field) susceptibility $\chi$ is defined by the dipole momentum $\langle D \rangle$ of the medium, induced by the probe field:

$$\langle D \rangle = \text{Tr} \left( \hat{\rho} \hat{d} \right) = \frac{F}{2}[e_p \chi \exp(-i\Omega t) + \text{c.c.}],$$

where $\hat{\rho}$ is the atomic density operator and $\hat{d}$ is the atomic dipole momentum operator. Although generally the susceptibility $\chi$ is a second rank tensor, for our geometry it is a scalar function. The atomic density operator obeys the Liouville equation with the Hamiltonian

$$\hat{H} = \hat{H}_0 - [f(t) + F(t)] \sum_{i,j} d_{ij} |i\rangle \langle j|,$$

where $\hat{H}_0$ is the field-free atomic Hamiltonian. The interaction operator of an atom with the radiation fields (in the dipole approximation) contains the double summation over the atomic
states, including the continuum states, and \( d_{ij} = \langle i | \hat{d} | j \rangle \) is the dipole matrix element. The Hamiltonian \( \hat{H}_0 \) includes the configuration interaction \( \hat{W} \), which causes autoionization of the discrete states \( a \) embedded in the continuum:

\[
\hat{H}_0 = \sum_i E_i |i\rangle \langle i| + \left( \sum_a \int W_{a,E_c} a \langle E_c | dE + \text{h.c.} \right),
\]

where the subscript \( c \) denotes the continuum atomic state with the energy \( E \). We neglect the radiation decay of the short-living \( (\sim 10^{-12} - 10^{-13} \text{ sec}) \) autoionizing states.

**Figure 1.** Scheme of transitions in He. The linearly polarized probe field (dashed arrows) scans the region of the AIS \( 2s2p^1P \), coupled by circularly polarized laser field (thick solid arrows) with the \( 2p^21S \) AIS (a), or the \( 2s3d^1D \) AIS (b). Horizontal arrows indicate autoionization into the adjacent continuum channels.

The schemes of resonant transitions in helium involved in the processes are shown in figure 1. We take the right circularly polarized coupling field \( \mathbf{e}_l = \mathbf{e}_{+1} = -(\mathbf{e}_x + i \mathbf{e}_y)/\sqrt{2} \). The acquired ellipticity (1) and the rotation angle (2) are considered for the VUV radiation in the region of the \( 2s2p^1P \) AIS in He, coupled with \( 2p^21S \) (figure 1a) or \( 2s3d^1D \) (figure 1b) AISs. We neglect weak two-electron transitions caused by the laser field from autoionizing states to the continuum below the second ionization threshold. Hereafter we indicate the ground state and the odd and even AISs by the labels “g”, “a”, and “b”, respectively. Our treatment is limited to a moderately strong coupling laser field with intensity \( I \leq 2 \cdot 10^{-4} \text{ a.u.} \) \( (1 \text{ a.u.} = 6.43 \cdot 10^{15} \text{ W/cm}^2) \), when multiphoton ionization from the ground state and coupling between the continuum states can be neglected. The Liouville equation for the density matrix elements \( \rho_{ij} \) is solved, within the rotating wave and adiabatic approximations, treating the probe field as a perturbation. After eliminating the continuum states [31] the set of differential equations for the modified density matrix elements \( \tilde{\rho}_{aM,g} = \rho_{aM,g} \exp(-i\Omega t) \) and \( \tilde{\rho}_{bM,g} = \rho_{bM,g} \exp(-i(\Omega + \omega)t) \) takes the form

\[
\begin{align*}
    i\dot{\tilde{\rho}}_{aM,g} &= (E_a - \Omega) \tilde{\rho}_{aM,g} - \Omega M \tilde{\rho}_{bM+1,g} - \frac{\pi T_a}{2} (q - i) \frac{F}{2} \tilde{\rho}_{E_a c M,g} \quad (7) \\
    i\dot{\tilde{\rho}}_{bM+1,g} &= (E_{bM+1} - \Omega) \tilde{\rho}_{bM+1,g} - \Omega M \tilde{\rho}_{aM,g} \cdot 
\end{align*}
\]

Here the (real) Rabi frequency is

\[
\Omega_M = \frac{f}{2} d_{bM+1,aM} = \sqrt{\frac{2\pi c}{f}} d_{bM+1,aM},
\]

\( \omega \)
where \( c \) is the speed of light and \( I \) is the laser intensity. The complex eigenenergies are defined by
\[
\mathcal{E}_a = E_a + \delta E_a - i\frac{\Gamma_a}{2}, \quad \mathcal{E}_{bM+1} = E_b + \delta E_b - \omega_L - E_g - i\frac{\Gamma_{bM+1}}{2},
\] (10)
where the widths and energy shifts are
\[
\Gamma_a = 2\pi |W_{E_a,c,a}|^2, \quad \delta E_a = \varphi \int \frac{|W_{E_a,c,a}|^2}{\Omega - E} dE,
\] (11)
\[
\Gamma_{bM+1} = 2\pi |W_{E_b,c,b}|^2 + 2\pi \sum_{c'} |d_{E'_c'M+2,bM+1}|^2 \frac{\gamma^2}{4}, \quad \delta E_b = \varphi \int \frac{|W_{E_b,c,b}|^2}{\Omega + \omega - E} dE. \tag{12}
\]
The label \( c' \) denotes the continuum states above the second ionization threshold with the energy \( E' = \Omega + 2\omega \) (with respect to the ground state). The corresponding term in \( \Gamma_{bM+1} \) describes the laser induced width of AIS “b”. In our calculations the influence of this term was found to be not important, and the shifts \( \delta E_{a(b)} \) are included in the AISs energy values.

The profile index of the AIS “a” in (7) is defined according to [32]
\[
q = \frac{d_{ag} + \varphi \int \frac{W_{a,E_c,d_{E_c,a}}}{\Omega - E} dE}{\pi W_{a,E_c,d_{E_c,g}}}
\] (13)
and is expressed, due to the Wigner-Eckart theorem, in terms of reduced dipole matrix elements, which are independent of the magnetic quantum numbers.

Noteworthy, for the elliptically polarized laser field, a unitary transformation of the atomic basis exists that reduces the equation set for density matrix elements to uncoupled pairs and singles of atomic superposition states [28, 33, 34, 35]. In the case of circular polarization such a reduction appears naturally. The solution of (7) and (8) for the density matrix element \( \tilde{\rho}_{cM,g} \) in the case of the two-level system has the form
\[
\tilde{\rho}_{aM,g} = \sqrt{\frac{\pi\Gamma_a}{2}} (q - i) \frac{F}{2} d_{E_a,cM,g} \frac{\Omega - \mathcal{E}_b}{(\Omega - \mathcal{E}_b)(\Omega - \mathcal{E}_a) - \Omega^2_M}. \tag{14}
\]
First we imply that the state “b” is the 2p\(^2\)1\(S\) AIS (figure 1a). Then the right polarized component of the probe field causes excitation of the uncoupled \( M = +1 \) sublevel of the state “a” and the corresponding solution of (7) is determined by
\[
\tilde{\rho}_{aM,g} = \sqrt{\frac{\pi\Gamma_a}{2}} (q - i) \frac{F}{2} d_{E_a,cM,g} \frac{1}{\Omega - \mathcal{E}_a}. \tag{15}
\]
The left circularly polarized component excites the sublevel \( M = -1 \) of “a”, which is coupled to the sublevel \( M = 0 \) of “b” and the corresponding solution is given by (14). Using the Wigner-Eckart theorem, one can express \( d_{0a,a-1} = d_{ba}/\sqrt{3} \). Then it follows from equations (4), (14), (15):
\[
\chi^{(-)} - \chi^{(+)} = \frac{\sigma_0 \Gamma_a}{8\pi k} (q - i)^2 \left( \frac{A_+}{\Omega - \mathcal{E}_+} + \frac{A_-}{\Omega - \mathcal{E}_-} - \frac{1}{\Omega - \mathcal{E}_a} \right), \tag{16}
\]
where \( \sigma_0 = 4\pi^2 k |d_{E_a,cM,g}|^2 \) is the direct photoionization cross section in the region of the AIS “a”. Note that, after eliminating the continuum, we must put \( d_{g,aM} = \sqrt{\frac{\pi \Gamma_a}{2}} (q - i) d_{g,E_a,cM} \) in (4).

The complex eigenenergies of the coupled sublevels and the corresponding complex amplitudes in equation (16) are
\[
\mathcal{E}_\pm = \frac{\mathcal{E}_a + \mathcal{E}_b}{2} \pm \frac{1}{2} \sqrt{(\mathcal{E}_a - \mathcal{E}_b)^2 + 4\Omega^2_M}, \tag{17}
\]
\[
\mathcal{A}_\pm = \pm \frac{\mathcal{E}_\pm - \mathcal{E}_b}{\mathcal{E}_+ - \mathcal{E}_-}. \tag{18}
\]
The real and imaginary parts of the complex eigenenergies (17) determine the position and width of resonances on the dispersion curve (16). The dependence of the complex eigenenergies (17) as functions of laser intensity $I$ and frequency detuning $\Delta = E_a - E_b + \omega$ were described in detail in [36].

For coupling of the AIS $2s2p^1P$ with the AIS $2s3d^1D$, being now the states “a” and “b”, respectively (figure 1b), both magnetic sublevels, $M = \pm 1$, of the state “a” are coupled with the corresponding magnetic sublevels of the state “b”. This gives, instead of (16), after straightforward transformations:

$$\chi^{(-)} - \chi^{(+)} = \frac{\sigma_0 \Gamma_a}{8 \pi k} (q - i) \sum_{h=\pm 1} (-h) \left( \frac{A_+^{(h)}}{\Omega - E_a^{(h)}} + \frac{A_-^{(h)}}{\Omega - E_b^{(h)}} \right),$$

(19)

where the complex eigenenergies and complex amplitudes for negative and positive probe field helicities $h$ are given by (17) and (18), with $d_{a-1,0} = d_{PD}/\sqrt{30}$ and $d_{a1,2} = d_{PD}/\sqrt{5}$, respectively.

At low laser intensity, equations (16) and (19) turn into

$$\chi^{(-)} - \chi^{(+)} = C \frac{\sigma_0 \Gamma_a}{4 k c} (q - i) |d_{ba}|^2 (\Omega - E_a) I,$$

(20)

where $C = \frac{1}{3}$ for the $1P-1S$ coupling and $C = \frac{-1}{6}$ for the $1P-1D$ coupling, showing the linear dependence of the optical activity on the laser field intensity.

3. Numerical results and discussion

The parameters of the involved helium states are partly known from accurate calculations and measurements [37, 38, 39, 40, 41]. We used the following values of the related parameters: $E_S = 62.068$ eV, $E_P = 60.136$ eV, $E_D = 63.864$ eV, $\Gamma_S = 5.87 \cdot 10^{-3}$ eV, $\Gamma_P = 3.73 \cdot 10^{-2}$ eV, $\Gamma_D = 5.44 \cdot 10^{-4}$ eV, $q = -2.8$. To find the lacking dipole matrix elements $d_{ab}$ we utilized the MCHF code [42]. Wave functions of the AISs $2s2p^1P$, $2p^21S$, and $2s3d^1D$ were calculated by diagonalization on a restricted basis of self-consistent configuration state functions with the corresponding LS symmetry. The nonorthogonality of the single electron radial wavefunctions for different AIS was taken into account in calculations of the matrix elements $d_{ab}$. The basis includes all two-electron configurations constructed from orbitals with principal quantum number $n = 2, 3, 4$, where the functions with $n = 4$ were pseudo orbitals, optimized on the energies of the corresponding AIS. Thus, the main part of the electron correlations was considered. The absolute energies of the three AISs of our interest were obtained with an accuracy of 0.1 eV, as compared with experiment and more precise calculations. The values of the reduced matrix elements were obtained as $d_{PS} = 3.64$ a.u. and $d_{PD} = 3.24$ a.u. The contribution of the laser induced width in (12) was found to be insignificant in our calculations.

Figures 2 and 3 show our results for the cases of $2s2p^1P - 2p^21S$ and $2s2p^1P - 2s3d^1D$ coupling, respectively. The absolute values of $\theta$ and $\epsilon$ correspond to the value of $NL = 5 \cdot 10^{16}$ cm$^{-2}$.

The surface plots in figures 2a and 3a present the dependence of the rotation angle $\theta$ on the probe field frequency $\Omega$ (the “dispersion function”) and on the laser field intensity at zero laser frequency detuning $\Delta$. The same plots in figures 2e and 3e exhibit the dependence of $\theta$ on the probe field frequency and the laser detuning at fixed laser intensity $I = 10^{14}$ W/cm$^2$.

Figures 2(b-d,f-h) and 3(b-d,f-h) show several cuts along the dispersion axis of the corresponding surface plots together with the dispersion functions for the acquired ellipticity $\epsilon$ and the photoabsorption cross section $\sigma$. At low laser intensity the cross section turns to the Fano profile of the He $2s2p^1P$ state [32, 40, 41]. The photoabsorption cross section does
Figure 2. Alteration of the VUV probe field polarization in the He medium \((NL = 5 \cdot 10^{16} \text{ cm}^{-2})\) due to resonant coupling of the \(2s2p^{1}P\) and \(2p^{2}\)\(^1\)S AISs by the laser field with right circular polarization. (a) Rotation angle variation with the probe photon energy and laser intensity. (b-d) Rotation angle (solid red line), acquired ellipticity (dashed blue line), and ionization cross sections (dashed-dotted line) spectral dependencies at zero detuning and different laser intensities: (b) \(I = 10^{10}\) W/cm\(^2\), (c) \(I = 10^{11}\) W/cm\(^2\), (d) \(I = 10^{12}\) W/cm\(^2\); (e) Rotation angle variation with the probe photon energy and laser detuning. (f-h) The same as in (b-d), but at the laser intensity \(I = 10^{11}\) W/cm\(^2\) and different values of the laser detuning: (f) \(\Delta = -\Gamma_P\), (g) \(\Delta = \Gamma_P\), (h) \(\Delta = 3\Gamma_P\). The cross sections are scaled on the left axes in units of \(10^{-16}\) cm\(^2\).
not exceed 10 Mb in the region of the AIS He 2s2p\(^1\)P even in the resonance maximum [43]. Therefore, the helium medium for the above value of \(NL\) is optically transparent. Note that the condition of weak nonlinearity, \(N|\chi| \ll 1\), is fulfilled. The parameters \(\epsilon\) and \(\theta\) sharply vary as functions of \(\Omega, \omega, \Delta, I\), and the rotation angle is up to 10°.

The general appearance of the dispersion functions for \(\epsilon\) and \(\theta\) in figures 2e and 3e resembles that for the discrete states in figures 2 and 3 of [11]. In our case, the symmetry of the dispersion curves for \(\epsilon\), as well as the antisymmetry for \(\theta\), are destroyed due to bound-free interference in

**Figure 3.** Same as in figure 2, but for coupling of the 2s2\(^1\)P and 2s3\(^1\)D AISs.
the AISs. At frequencies $\Omega$ where the acquired ellipticity steeply crosses zero, the rotation angle assumes maximum values due to the Kramers-Kronig relation. The positions of the maxima in the dispersion functions are shifted in such a way that one could obtain maximum polarization rotation with small acquired ellipticity or maximum ellipticity with small rotation.

For the $^1P - ^1S$ coupling (figure 1a), equation (16) contains three resonant terms with the complex coefficients. Hence, the dispersion functions for the rotation angle and the acquired ellipticity, as well as the photoionization cross section, generally show three asymmetric resonances, which can overlap (figure 2). The positive helicity component of the probe field produces the excitation of the $2s2p^1PM = 1$ sublevel that is independent on the laser parameters. The outermost two resonances are produced by the negative helicity component of the probe field and constitute an Autler-Townes doublet [44], originating from the ladder scheme in figure 1a. With increasing of the laser intensity, the widths of the two resonances tend to the mean value $(\Gamma_a + \Gamma_b)/2$ [36] and their energies deviate from the mean value $\frac{1}{2}(E_S + E_P - \omega)$ roughly as $\pm d_{SP}\sqrt{I}$.

Manipulating the laser detuning yields another control mechanism for the optical activity. As demonstrated in figures 2(e-h), the values of $\epsilon$ and $\theta$ change drastically when the laser frequency scans the region of the AIS $2s2p^1P$, due to shifting of the Autler-Townes doublet with respect to the central resonance. Note that the controlling efficiency of the laser detuning is restricted by the fact that the coupling strength reduces when the detuning increases.

For the $^1P - ^1D$ coupling (figures 1b, 3), equation (19) contains four resonant terms, leading to four generally overlapping resonance structures in the dispersion functions. In contrast to the $^1P - ^1S$ coupling, there is no “reference” resonance unaffected by the optical coupling: the resonances of the both doublets depart with increasing laser intensity (figures 3(a-d)). The laser detuning shifts the resonance positions to one side as well (figure 3e). Nevertheless, it is possible to obtain an established value of nonlinearity in a narrow frequency region using both laser intensity and detuning.

4. Conclusion
We have studied theoretically the optical activity of an atomic medium in the spectral region of the $2s2p^1P$ autoionizing state in He coupled by a circular field with the $2p^2S$ or $2s3d^1D$ states. Analytical expressions and numerical results were obtained for the laser induced polarization rotation angle and the acquired ellipticity of the linearly polarized probe VUV radiation. A rotation angle of $\sim 10^\circ$ can be obtained for optically transparent helium media at laser intensities of $10^{10} - 10^{12}$ W/cm$^2$. The optical activity in the VUV region can be efficiently controlled by manipulating the intensity and frequency of the coupling optical field.

The present investigation may be extended to other targets and states, including core-hole autoionizing hole and Auger states. Laser coupling of the Auger states will possibly lead to laser induced optical activity of a medium in the X-ray wavelength range.

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