

# Angular Momentum Transfer at the Chemo-ionization and Spin Exchange Processes. Interaction between Spin-Polarized Metastable Helium Atoms

Victor Kartoshkin

Ioffe Institute, Politeknicheskaya ul. 26, St. Petersburg, 194021, Russia

**Abstract:** In the collision of spin-polarized atoms, the transmission of electron polarization between colliding particles is possible. A similar situation occurs in a collision, for example, between alkali atoms in the ground state. Suppose one of the colliding particles is excited, and its energy is sufficient for the ionization of the collision partner, then along with the elastic process (spin exchange). In that case, an inelastic process (ionization) is also possible; as a result, polarization can also be transferred to the partner. Simultaneously occurring elastic and inelastic processes at the collision between two helium atoms are possible if these atoms are in the metastable ( $2^3S_1$ ) state. The paper discusses the interaction between spin-polarized metastable atoms of helium-3 and helium-4 isotopes under the condition of optical pumping of atoms. Complex interaction potentials of the atoms under consideration were constructed, and on their basis, the spin exchange and chemo-ionization cross-sections were calculated. Using these cross-sections, expressions for the transfer of orientation and alignment from one atom to another were obtained.

**Keywords:** Ionization; Polarization transfer; Angular moment.

## 1. Introduction

The study of interaction involving spin-polarized atoms is possible in experiments on the optical orientation of atoms. The optical orientation of atoms is the transfer of angular momentum from polarized resonant radiation to an ensemble of atoms that are either in the ground state or in an excited state and have an uncompensated electron spin. As an example of the optical orientation of atoms in the ground state, we can consider the optical orientation of alkali atoms. Atoms of inert gases in the ground state are diamagnetic. At the same time, in excited (metastable) states, these atoms are paramagnetic and, therefore, can also be polarized. Experiments of this kind were carried out on He, Ne, Ar, Xe, and Kr atoms<sup>1,2</sup>. Since metastable helium atoms have an electron spin of  $S = 1$ , both oriented and aligned atoms can be obtained in the process of optical orientation. Aligned metastable helium atoms can be obtained by irradiating an ensemble of atoms with linearly polarized resonant radiation, and oriented metastable helium atoms are obtained by interacting with circularly polarized radiation. In the second case, alignment also occurs in the system.

Even though studies of ionization processes involving triplet metastable helium atoms began quite a long time ago, they have not lost their relevance in our

time. In there were presented very accurate theoretical results<sup>3</sup> of Penning ionization rate coefficients of the excited metastable helium atoms ( $^4\text{He}(2^3S)$  and  $^3\text{He}(2^3S)$ ) colliding with the hydrogen isotopologues ( $\text{H}_2$ , HD,  $\text{D}_2$ ) in the ground and first excited rotational and vibrational states.

Investigation of the spin-dipole mediated interactions between metastable helium atoms in the spin-polarized quintet  $^5\Sigma_g^+$  state to the singlet  $^1\Sigma_g^+$  state from which autoionization is highly probable, resulting in finite lifetimes for the atoms in a trap<sup>4</sup>.

At the interaction of metastable polarized helium atoms in the  $2^3S_1$ -state with helium atoms in the ground state, a well-known process of metastable exchange occurs<sup>5</sup>. As a result of this process, excitation is transferred from the metastable atom to the atom in the ground state<sup>6</sup>. Furthermore, if the excited atom was preliminarily polarized, then along with the transfer of excitation, the transfer of polarization also occurs. Possibilities of this kind are used to obtain nuclear-polarized atoms of the 3-He isotope, which are used in medicine for nuclear tomography<sup>7</sup>.

The metastable helium atom ( $\text{He}^*$ ), having a large internal energy store (19.8 eV), can ionize the molecules or atoms even at the thermal energies of relative motion. In a collision between two excited

\*Corresponding author: Victor Kartoshkin

Email address: [victor.kart@mail.ioffe.ru](mailto:victor.kart@mail.ioffe.ru)

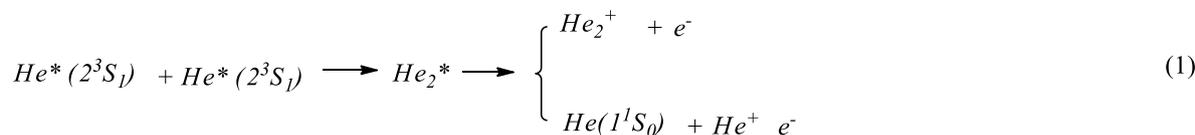
DOI: <http://dx.doi.org/10.13171/mjc02210071641kartoshkin>

Received June 30, 2022

Accepted September 6, 2022

Published October 7, 2022

helium atoms in the metastable triplet state, the chemo-ionization process takes place<sup>8,9</sup>. A metastable helium atom has electron spin  $\mathbf{S} = \mathbf{1}$ , and at the collision between two metastable helium atoms, two simultaneous processes are possible. These two processes are chemo-ionization (1) and spin exchange (2). This situation is typical for collisions of polarized



As noted above, simultaneously with the inelastic



where  $m_1$  and  $m_2$  are the projections of the angular moments of the atomic particles.

If collisional atoms are polarized in spin, then angular momentum transfer from one atom to another is possible in processes (1) and (2). It should be borne in mind that the simultaneous occurrence of elastic and inelastic processes leads to their mutual influence. Although, as shown below, these two processes significantly affect each other in particular, the cross-sectional view of the elastic process changes significantly. This is the case for both the cross-section of the spin exchange and the frequency shift cross-section.

## 2. Elastic and inelastic processes at the collisions between spin-polarized atomic particles

At the collision of polarized atomic particles with uncompensated electron spin, if at least one of them has energy sufficient to ionize the collision partner, then along with the elastic process (in this case, spin exchange), the process of ionization of the collisional partner is also possible. In the case of a collision of  $f_{q,t,s} = \frac{1}{2ik} \sum_{l=0}^{\infty} (2l+1) [\exp(2i\eta_l^{q,t,s}) - 1] P_l(\cos\theta)$

Where the phase shifts ( $\eta_l^{q,t,s}$ ) can be extracted from

$$\frac{d^2 g_l^{q,t,s}}{dR^2} + \left[ k^2 - V_{q,t,s} - \frac{l(l-1)}{R^2} \right] g_l^{q,t,s} = 0 \quad (4)$$

behaves asymptotically as

$$g_l^{q,t,s} \sim k^{-1} \sin\left(kR - \frac{1}{2}l\pi + \eta_l^{q,t,s}\right) \quad (5)$$

Here  $l$  is the orbital moment,  $k$  is the wave vector,  $V_{q,t,s}$  is the quintet, triplet, or singlet interaction potentials,  $P_l$  is the Legendre polynomials.

As noted above, both spin exchange and ionization are possible for singlet and triplet terms, while only spin exchange is possible for quintet terms. Therefore, the scattering phase is real for the quintet term and complex for the triplet and singlet terms. On the other hand, for the term with the greater multiplicity  $V_q$ , the ionization process (1) is forbidden in accordance with the Wigner spin conservation rule<sup>13</sup> since the total spin of the initial species is 2, whereas that of products of the reaction (1) is 1. Also, due to low collision time,

metastable helium atoms with alkali metal atoms, for example, with sodium atoms in the ground state<sup>10</sup>.

In a collision of two triplet metastable helium atoms, a quasi-molecule  $He_2^*$  is formed<sup>11</sup>. This quasi-molecule can decay in two channels, with the formation of atomic and molecular helium ions.

process (1) an elastic process is also possible:

two excited metastable helium atoms in the  $2^3S_1$  state, having internal energy of 19.8 eV and an electron spin  $\mathbf{S}_1 = \mathbf{S}_2 = \mathbf{1}$ , both an elastic process, i.e., the transfer of electron polarization from one atom to another (this is a well-known spin exchange process) and an inelastic process (the ionization process of a metastable helium atom) are possible. The quasi-molecule formed in such a collision can be in one of three states with a total spin  $S = 0, 1, \text{ and } 2$ , corresponding to three molecular terms  $^1\Sigma^+, ^3\Sigma^+, ^5\Sigma^+$  (singlet, triplet, and quintet).

Following the law of conservation of total spin ( $m_1 + m_2 = m_1' + m_2'$ ), ionization is only possible from the states of low multiplicity, i.e., from the singlet and triplet states, since, in this case, the total spin is the same at the exit and entrance of the reaction.

The scattering amplitude on singlet ( $f_s$ ), triplet ( $f_t$ ), and quintet ( $f_q$ ) terms may be expressed in terms of the scattering phase ( $\eta_l^{q,t,s}$ )<sup>12</sup>:

$$f_{q,t,s} = \frac{1}{k} \sum_{l=0}^{\infty} (2l+1) [\exp(i\eta_l^{q,t,s}) - 1] P_l(\cos\theta) \quad (3)$$

the regular solutions of the partial wave equation:

$$\frac{d^2 g_l^{q,t,s}}{dR^2} + \left[ k^2 - V_{q,t,s} - \frac{l(l-1)}{R^2} \right] g_l^{q,t,s} = 0 \quad (4)$$

there are insufficient transitions between doublet and quartet terms of the quasi-molecule ( $He_2^*$ ), which allows ignoring the influence of the spin exchange process on chemo-ionization. Hence, we might put the phase shifts to be real for the quartet term ( $\eta^q$ ) and complex for the triplet and singlet terms ( $\eta^{t,s} = \chi^{t,s} + i\lambda^{t,s}$ ).

The chemo-ionization cross-sections are as follows<sup>12</sup>:

$$\sigma_{t,s}^{abs} = \pi/k^2 \sum_{l=0}^{\infty} (2l+1) [1 - \exp(-4\lambda_l^{s,t})] \quad (6)$$

Following earlier works<sup>14,15</sup>, the spin exchange cross-section can be defined as follows. Let

$\Phi_{\text{HeHe}}(S_1, m_1; S_2, m_2)$  be the initial (just before collision) wave function of the  $(\text{He}_2)^*$  system ( $S_1$  and  $S_2$  are the electron spins of the metastable helium atoms, and  $m_1$

and  $m_2$  are the projections of spins). The asymptotic wave function (after scattering) will be as

$$\Psi_{\text{He}^*\text{He}^*}(S_1 m_1; S_2 m_2) \sim \Phi_{\text{He}^*\text{He}^*}(S_1 m_1; S_2 m_2) \cdot \exp(ikr) + \frac{\exp(ikr)}{r} \sum_{m_1 m_2} \Phi_{\text{He}^*\text{He}^*}(S_1 m_1; S_2 m_2) \cdot f(m_1 m_2; m'_1 m'_2; \theta) \quad (7)$$

Where  $k$  is the initial wave vector, the vector  $r$  describes the relative position of nuclei, and  $f(m_1 m_2; m'_1 m'_2; \theta)$  is the scattering amplitude through the angle  $\theta$  in the system of the center of mass.

According to the other angular moments coupling scheme, the spins  $S_1$  and  $S_2$  should give the total spin  $S$  with the values 0, 1 or 2 and the magnetic quantum number  $M_S$ , which corresponds to the molecular wave function  $\Xi(S, M_S)$ , that yields

$$\Psi_{\text{He}^*\text{He}^*}(S_1 m_1; S_2 m_2) \sim \sum_S C_{S_1 m_1 S_2 m_2}^{SM_S} \Xi(S, M_S) \cdot \left[ \exp(ik \cdot r) + f_s(\theta) \frac{\exp(ik \cdot r)}{r} \right] \quad (8)$$

here  $M_S = m_1 + m_2$ ,  $C_{S_1 m_1 S_2 m_2}^{SM_S}$  the Clebsch – Gordan coefficients, and  $f_s(\theta)$  is the amplitude of elastic scattering for the singlet, triplet, or quintet term.

From (7) and (8), one can conclude that the amplitude of changing the atom's spin coordinates should be connected with the scattering amplitudes on molecular terms through the vector coupling coefficients as

$$f(m_1 m_2; m'_1 m'_2; \theta) = \sum_S C_{S_1 m_1 S_2 m_2}^{SM_S} C_{S_1 m'_1 S_2 m'_2}^{S'M'_S} f_S(\theta) \quad (9)$$

The cross-section of the spin exchange process  $m_1 m_2 \rightarrow m'_1 m'_2$  when scattering by the angle  $\theta$  is then given by the expression:

Using the Clebsch – Gordan coefficients values for the angular moments  $S_1=1$  and  $S_2=1$  leads to the conclusion that in such a case, all cross-sections for spin exchange of collision partners should be the following:

$$\sigma(m_1 m_2; m'_1 m'_2; \theta) = |f(m_1 m_2; m'_1 m'_2; \theta)|^2 \quad (10)$$

$$\sigma_1(\theta) = \sigma(1, 0; 0, 1) = \sigma(0, 1; 1, 0) = \sigma(-1, 0; 0, -1) = \sigma(0, -1; -1, 0) = \frac{1}{4} |f_t - f_q|^2$$

$$\sigma_2(\theta) = \sigma(1, -1; 0, 0) = \sigma(0, 0; 1, -1) = \sigma(-1, 1; 1, -1) = \sigma(0, 0; 0, 1) = \frac{1}{9} |f_s - f_q|^2 \quad (11)$$

$$\sigma_3(\theta) = \sigma(1, -1; -1, 1) = \sigma(-1, 1; 1, -1) = \left| \frac{1}{3} f_s - \frac{1}{2} f_t - \frac{1}{6} f_q \right|^2$$

Thus,  $\sigma_1$  is determined by the interference of elastic scattering amplitudes on triplet and quintet terms,  $\sigma_2$  on singlet and quintet terms, and  $\sigma_3$  on all three terms. Integration concerning the angle  $\theta$ , taking into

account expression (3), leads us to the following form of a complete cross-section of spin exchange at the collision of polarized metastable helium atoms (for example, for the cross-section  $\sigma_1^{tr}$ ):

$$\sigma_1^{tr} = 2\pi/4 \int_0^\pi |f_t - f_q|^2 = \pi/k^2 \sum_{l=0}^\infty (2l+1) [1 - 2\exp(-2\lambda_l^t) \cos(\chi_l^q - \eta_l^d) + \exp(-4\lambda_l^t)] \quad (12)$$

similarly, using (11) one can obtain expressions for the spin exchange cross sections  $\sigma_2^{tr}$  and  $\sigma_3^{tr}$ .

$$\sigma_1^{tr} = \pi/4k^2 \sum_{l=0}^\infty (2l+1) [1 - 2\exp(-2\lambda_l^t) \cos(\chi_l^t - \eta_l^q) + \exp(-4\lambda_l^t)] \quad (13)$$

$$\sigma_2^{tr} = \pi/9k^2 \sum_{l=0}^\infty (2l+1) [1 - 2\exp(-2\lambda_l^s) \cos 2(\chi_l^s - \eta_l^q) + \exp(-4\lambda_l^s)] \quad (14)$$

$$\sigma_3^{tr} = \frac{\pi}{36k^2} \sum_{l=0}^\infty (2l+1) [1 - 12 \exp(-2\lambda_l^s - 2\lambda_l^t) \cos 2(\chi_l^s - \chi_l^t) + 4 \exp(-2\lambda_l^s) \cos 2(\eta_l^q - \chi_l^s) - 6 \exp(-2\lambda_l^t) \cos 2(\eta_l^q - \chi_l^t) + 4 \exp(-4\lambda_l^s) + 9 \exp(-2\lambda_l^t)] \quad (15)$$

The above formulas  $\lambda_l^{s,t}$  and  $\chi_l^{s,t}$  are the imaginary and real parts of the complex phase ( $\eta_l^q = \chi_l^{s,t} + i\lambda_l^{s,t}$ ) of scattering on the singlet and triplet terms of the quasi-molecule.  $\eta_l^q$  is the scattering phase on the quintet term.

scattering phases on a singlet, triplet, and quintet potentials.

### 3. Complex potentials

Thus, to consider the transfer of polarization in the collision between metastable helium atoms, one needs to know the values of the cross-sections of spin exchange and chemo-ionization, i.e., following (13)-(15) and (6), one can calculate the

Collisions of two metastable helium atoms can be described using complex interaction potentials of the  $\text{He}_2^*$  quasi-molecule. Since the metastable helium atoms in the  $2^3S_1$ -state have the electron spin  $S = 1$ , in the process of their interaction, a quasi-molecule is formed in the states  $^1\Sigma^+$ ,  $^3\Sigma^+$ ,  $^5\Sigma^+$  (corresponding to the total spin  $S = 0, 1$ , and  $2$ ). The ionization process

is possible only in states with lower multiplicity, i.e., in the singlet and triplet states; then, these states can be described by a complex interaction potential of the form

$$V(\mathbf{R}) = U(\mathbf{R}) - \frac{i}{2}\Gamma(\mathbf{R}) \quad (16)$$

Where the real part of the potential ( $U(R)$ ) describes the scattering of particles upon collision, and the imaginary part ( $\Gamma(R)$ ) describes the destruction of particles in the ionization process.

The process of ionization (Penning, associative, etc.) causes a decrease in the number of particles compared to the number of them that have entered the reaction. This particle absorption can be described phenomenologically by introducing the imaginary part of the potential energy, i.e., based on the integrated interaction potential (16).

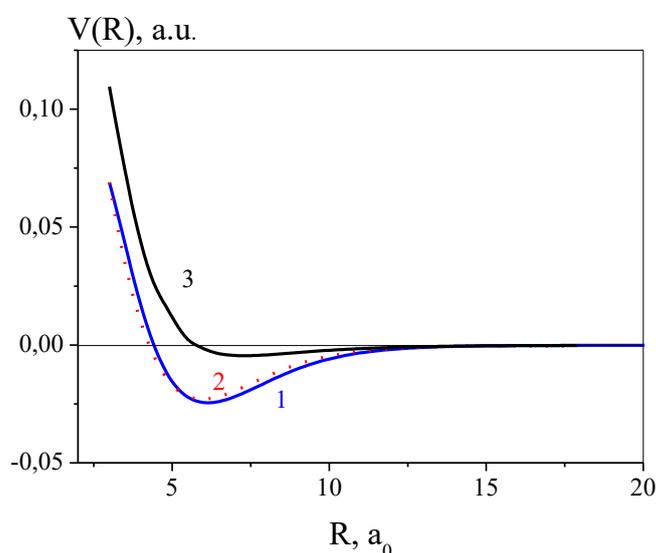
As it is known, in optics, the complex reflection coefficient is introduced to describe the reflection of light from an absorbing medium. For this reason, the imaginary part of the complex potential (16), which is quite similar to the width of an atomic level with a finite lifetime, is called the potential width or autoionization width. The complex or optical potential method has become widespread in nuclear and molecular physics and is applied to describe the chemo-ionization process<sup>16,17</sup>.

At the same time, the quartet state of a quasi-molecule can be described by the real interaction potential of the form

$$V(\mathbf{R}) = U(\mathbf{R}) \quad (17)$$

As noted above, the interaction of two metastable atoms of helium atoms with electron spins  $\mathbf{S}_{1,2} = \mathbf{1}$  results in the formation of the  $\text{He}_2^*$  quasimolecule, which can be described by three interaction potentials  $V_s$ ,  $V_t$ , and  $V_q$ , corresponding to the total  $\mathbf{S} = \mathbf{0}, \mathbf{1}, \mathbf{2}$ . For singlet and triplet terms, both elastic scattering (in our case, spin exchange (2)) and absorption of the original particles (chemo-ionization (1)) are possible. However, for a quintet term, only elastic scattering is possible. This is because, in the case of the process (1) for the quintet term, the Wigner rule<sup>13</sup> would be violated, i.e., the law of conservation of the total spin  $\mathbf{S}$  during the reaction (1) (for the initial products, the total momentum was  $\mathbf{2}$ , and for the reaction products only  $\mathbf{1}$ ). On the other hand, the Wigner rule holds well for particles with zero orbital momentum, which is confirmed by experiments on the optical orientation of atoms<sup>1</sup>. Thus, the singlet and triplet terms of the  $\text{He}_2^*$  quasimolecule can be represented in the form (16), and the quintet term in the form (17). As usual, the autoionization width can be represented as an exponent function.

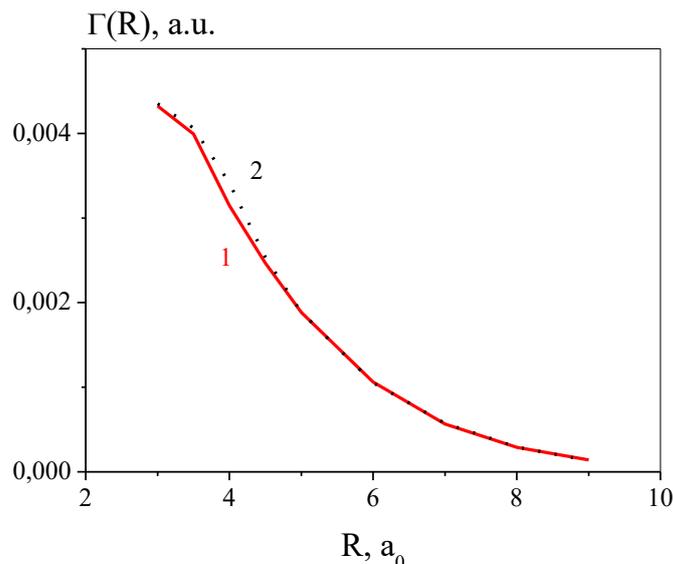
Figure 1 shows the real parts ( $U(R)$ ) of the potentials of a quasi-molecule in the singlet, triplet, and quartet states<sup>16</sup>.



**Figure 1.** Real parts of the interaction potentials of the  $\text{He}_2^*$  quasi-molecule. ((1) is the  $^1\Sigma^+$  state, (2) is the  $^3\Sigma^+$  state, and (3) is the  $^5\Sigma^+$  state; the distance  $R$  is given in Bohr radii)

Figure 2 shows the imaginary parts ( $\Gamma(R)$ ) of the

singlet and triplet terms (autoionization widths)<sup>16</sup>.



**Figure 2.** Imaginary parts of the interaction potentials of the  $He_2^*$ quasi-molecule. ((1) is the  $^1\Sigma^+$  state, (2) is the  $^5\Sigma^+$  state, and the distance R is given in Bohr radii)

Thus, using the interaction potentials of the  $He_2^*$  quasi-molecule together with the expressions for the cross-sections (13) - (15) and (6), one can calculate the cross-sections.

#### 4. Redistribution of spin polarization at the collision of polarized metastable helium atoms

The interaction of two metastable polarized helium atoms can be considered in the approximation of the density matrix. For example, the following equation can describe the evolution of the density matrix of atom A:

$$\frac{d^{(A)}}{dt}\rho = \left[ \left( \frac{\partial}{\partial t} \right)_{s.e.} + \left( \frac{\partial}{\partial t} \right)_{c.i.} + \left( \frac{\partial}{\partial t} \right)_{dif} + \left( \frac{\partial}{\partial t} \right)_{light} \right] (18)$$

Here, the first term describes a change in the density matrix due to spin-exchange collisions of atoms A and B. The second term represents a change in the density matrix due to chemo-ionization in collisions between atoms A and B. The third term describes a change in the density matrix because of collisions of atoms A with other particles in a gas discharge. The fourth term corresponds to the influence of diffusion on the

absorption-chamber walls. Finally, the last term corresponds to the influence of pumping light.

Taking into account the first two terms in equation (18), it is possible to move from the equation for the density matrix to the equations for the observed experiment values. These values are the longitudinal orientation and alignment: where  $\langle S_A \rangle_z$  is the longitudinal orientation of the particle A [ $\langle S_A \rangle_z = (\rho_{11} - \rho_{33})$ ],  $\langle S_B \rangle_z$  is the longitudinal orientation of the particle B [ $\langle S_B \rangle_z = (P_{11} - P_{33})$ ],  $\langle Q_B \rangle^{zz}$  is the longitudinal components of the electronic alignment of the particle B [ $\langle Q_B \rangle^{zz} = \frac{1}{2}(P_{11} + P_{33}) - P_{22}$ ]. Here  $P_{ii}$  and  $\rho_{ii}$  are the diagonal matrix elements of the density matrices  $P$  and  $\rho$ , describing atoms A and B.

Kinetic equations for the longitudinal components of orientation and alignment have the form

$$\begin{aligned} \frac{d\langle S_A \rangle_z}{dt} = & -N_B v \left[ \left( \frac{1}{3}\sigma_1^{tr} + \frac{1}{3}\sigma_2^{tr} + \frac{2}{3}\sigma_3^{tr} \right) + \left( \frac{1}{9}\sigma_s^{abs} + \frac{1}{3}\sigma_t^{abs} \right) \right] \langle S_A \rangle_z + N_B v \left[ \left( \frac{1}{3}\sigma_1^{tr} + \frac{1}{3}\sigma_2^{tr} + \frac{2}{3}\sigma_3^{tr} \right) + \right. \\ & \left. + \left( \frac{1}{9}\sigma_s^{abs} + \frac{1}{6}\sigma_t^{abs} \right) \right] \langle S_B \rangle_z + N_B v \left[ \left( \frac{1}{3}\sigma_1^{tr} - \frac{1}{6}\sigma_2^{tr} - \frac{1}{3}\sigma_3^{tr} \right) + \left( -\frac{1}{18}\sigma_s^{abs} + \frac{1}{12}\sigma_t^{abs} \right) \right] \langle S_A \rangle_z \langle Q_B \rangle^{zz} + \\ & + N_B v \left[ \left( -\frac{1}{3}\sigma_1^{tr} + \frac{1}{6}\sigma_2^{tr} + \frac{1}{3}\sigma_3^{tr} \right) + \left( \frac{1}{18}\sigma_s^{abs} + \frac{1}{12}\sigma_t^{abs} \right) \right] \langle S_B \rangle_z \langle Q_A \rangle^{zz} \end{aligned} \quad (19)$$

$$\begin{aligned} \frac{d\langle Q_A \rangle^{zz}}{dt} = & -N_B v \left[ (\sigma_1^{tr} + \sigma_2^{tr}) + \left( \frac{1}{9}\sigma_s^{abs} + \frac{1}{3}\sigma_t^{abs} \right) \right] \langle Q_A \rangle^{zz} + N_B v \left[ (\sigma_1^{tr} + \sigma_2^{tr}) + \left( -\frac{1}{9}\sigma_s^{abs} + \frac{1}{6}\sigma_t^{abs} \right) \right] \langle Q_B \rangle^{zz} + \\ & + N_B v \left[ \frac{3}{2}\sigma_2^{tr} + \left( \frac{1}{6}\sigma_s^{abs} + \frac{1}{4}\sigma_t^{abs} \right) \right] \langle S_A \rangle_z \langle S_B \rangle_z + N_B v \left[ \frac{1}{2}\sigma_2^{tr} + \left( \frac{1}{18}\sigma_s^{abs} - \frac{1}{12}\sigma_t^{abs} \right) \right] \langle Q_A \rangle^{zz} \langle Q_B \rangle^{zz} \end{aligned} \quad (20)$$

The explicit form of the cross-sections  $\bar{\sigma}_i^{tr}$  and  $\bar{\sigma}_{s,t}^{abs}$  are represented by the expressions (6) and (13) – (15). Here  $N_{A,B}$  is the concentration of the particles A and B,  $v$  is the relative thermal velocity of atoms.

Consider expressions (19) and (20). Square brackets in expressions (19) and (20) characterize the change in the orientation of atom A as a result of its collision with atom B. In this case, the sign (-) before the bracket indicates the loss of orientation of the ensemble of atoms A during the collision. The sign (+) before the bracket corresponds to the acquisition of orientation by atom A. Parentheses, which include sections with the index tr, are associated with the spin exchange process. The chemo-ionization process determines those that have sections with the index abs. The last two square brackets in (19) and (20) are connected with the influence of alignment  $\langle Q \rangle$  on the change in the orientation of atoms A in the processes

$$\begin{aligned}\sigma_{or}^{tr} &= \sigma_1^{tr}/3 + \sigma_2^{tr}/3 + 2\sigma_3^{tr}/3, \\ \sigma_{or}^{abs} &= \sigma_s^{abs}/9 + \sigma_t^{abs}/3, \\ \sigma_{al}^{tr} &= \sigma_1^{tr} + \sigma_2^{tr} \\ \sigma_{al}^{abs} &= \sigma_s^{abs}/9 + \sigma_t^{abs}/9\end{aligned}\quad (21)$$

Here, the subscripts “or” and “al” refer to the orientation and alignment,  $s$  and  $t$  denote singlet and triplet terms. Below there are the results of calculating the chemo-ionization and spin exchange cross-sections.

$$\eta_l = \int_{R_0}^{\infty} F_1(R) dR - \int_{R_0'}^{\infty} F_0(R) dR \quad (22)$$

where

$$\begin{aligned}F_1^S(R) &= \left[ 2\mu(E - V_S(R) - \frac{(l+1/2)^2}{2\mu R^2}) \right], \\ S &= s, t, q, \\ F_0(R) &= \left[ 2\mu E - \frac{(l+1/2)^2}{R^2} \right].\end{aligned}\quad (23)$$

Here,  $E$  is the kinetic energy,  $R_0$  and  $R_0'$  are the roots of the equations  $F_1^S(R) = 0$  and  $F_0(R) = 0$  (the larger root is taken for  $F_1^S$ ), and  $V_S(R)$  is the interaction potential corresponding to the total spin  $S$ .

The scattering phases obtained in this way were used to calculate the cross-sections of spin exchange and chemo-ionization following the ratios (6) and (13)-(15). This system's cross-sections were calculated earlier, considering the slight difference between singlet and triplet terms<sup>12</sup>. In this case, the cross-sections of spin exchange and chemo-ionization

of spin exchange and chemo-ionization. Since the value of orientation and alignment in experiments on the optical orientation of helium atoms, as usual, is not large ( $\leq 10\%$ ), the terms with the product  $\langle S \rangle \langle Q \rangle$  can be neglected.

The destruction of the orientation of A atoms as a result of spin exchange and chemo-ionization collisions with atoms B is determined by the first square bracket in equation (19). The destruction of the alignment of A atoms as a result of spin exchange and chemo-ionization collisions with atoms B is determined by the second square bracket in equation (19). Therefore, we can introduce the cross-sections  $\sigma_{or,al}^{abs}$  and  $\sigma_{or,al}^{tr}$ , which, according to Eqs. (19) - (20), characterize the destruction and transfer of orientation and alignment of colliding particles as a result of chemo-ionization and spin exchange processes as follows:

The scattering phases were determined by the Jeffrey's approximation modified by Langer<sup>7</sup> in the collision energy range of  $E = 10^{-2} - 10^{-4}$  a.u.:

included in equations (19) and (20) can be represented as

$$\begin{aligned}\sigma_s^{abs} &= \sigma_t^{abs} = \sigma^{abs} \\ 4\sigma_1^{tr} &= 9\sigma_2^{tr} = 36\sigma_3^{tr} = \sigma^{tr}\end{aligned}\quad (24)$$

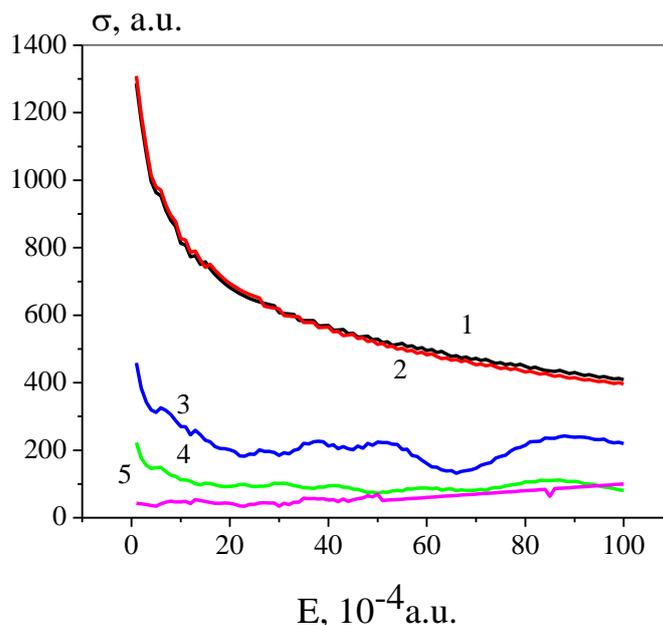
Table 1 shows the calculation results and Maxwell averaging of the cross-sections  $\sigma^{tr}$  and  $\sigma^{abs}$ . The cross-sections were calculated in distances between atoms from 3 to 15  $a_0$ . The collisional energies were from  $E = 10^{-2}$  to  $6 \cdot 10^{-3}$  a.u.

**Table 1.** Spin exchange  $\sigma^{tr}$  and chemo-ionization  $\sigma^{abs}$  cross-sections for collisions of helium isotopes  $^3\text{He}$  and  $^4\text{He}$ , taking into account (24) <sup>17</sup>.

T,K	50	100	150	200	250	300	350	400
$\sigma^{tr}, 10^{-16} \text{cm}^2$	25,2	40,1	45,7	48,5	49,9	50,6	50,7	50,2
$\sigma^{abs}, 10^{-16} \text{cm}^2$	115,8	151,5	160,8	162,6	161,0	157,6	152,8	147,2

Without considering the equality of singlet and triplet interaction potentials, the cross-sections included

in (19) and (20) are shown in Figure 3.

**Figure 3.** Energy dependencies of the spin exchange and chemo-ionization cross-sections

1-  $\sigma_1^{abs}$ , 2 -  $\sigma_2^{abs}$ , 3 -  $\sigma_1^{tr}$ , 4 -  $\sigma_2^{tr}$ , 5 -  $\sigma_3^{tr}$ . (The collisional energy  $E$  is given in atomic units, and the cross-section  $\sigma$  is given in atomic units).

The cross-sections were calculated in distances between atoms from 3 to 30  $a_0$ . The collisional energies were from  $E = 10^{-2}$  to  $10^{-4}$  a.u. Figure 3 shows the dependences of the spin exchange ((3)-(5)), and chemo-ionization ((1),(2)) cross sections calculated by the relations (13)-(15) and (6) using interaction potentials of the form (16) and (17). The chemo-ionization cross-sections were calculated using standard formulas for the chemo-ionization cross-section (6). These cross-sections characterize the destruction of metastable atoms in a collision. In the case of a collision of two metastable atoms, the

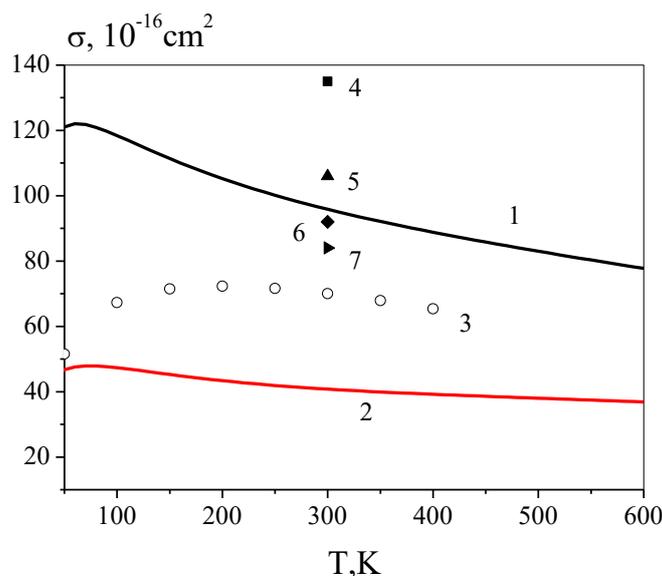
$$\sigma(T) = \frac{\langle \sigma(E)v \rangle}{\langle v \rangle} = \frac{1}{(kT)^2} \int_0^\infty \sigma(E) E \exp(-E/kT) dE \quad (25)$$

Here  $E$  is the collisional energy,  $k$  is the Boltzmann constant,  $T$  is the temperature,  $\sigma(E)$  and  $\sigma(T)$  are the dependences of the cross sections on the energy and temperature, and  $v$  is the relative velocity of colliding particles.

spin-exchange cross sections differ significantly from when the spin-exchange process is not accompanied by chemo-ionization. The simultaneous occurrence of elastic and inelastic processes leads to the spin exchange cross-section depending on the inelastic process. This manifests itself in the presence in expressions (13)-(15) of the imaginary parts of the complex scattering phase ( $\eta_{t,s} = \chi_{t,s} + i\lambda_{t,s}$ ). Since the real and imaginary parts of the singlet and triplet potentials are close in value<sup>16</sup>, the chemo-ionization cross sections are also close.

In order to get the cross sections included in equations (19) and (20), it is necessary to carry out the Maxwellian averaging of the presented in Figure 3 cross sections:

The destruction of a metastable helium atom's orientation due to chemo-ionization and spin exchange processes occurs with the cross sections, which are determined by expressions (21) ( $\sigma_{or}^{abs}$  and  $\sigma_{or}^{tr}$ ).



**Figure 4.** The spin-exchange ( $\sigma_{or}^{tr}$ ) and chemo-ionization ( $\sigma_{or}^{abs}$ ) cross-sections are dependent on temperature

- |  |  |
|--|--|
| 1- $\sigma_{or}^{abs}$ (this work)                 | 5 - $\sigma_{or}^{abs}$ (from work <sup>16</sup> ) |
| 2 - $\sigma_{or}^{tr}$ (this work)                 | 6 - $\sigma_{or}^{abs}$ (from work <sup>19</sup> ) |
| 3 - $\sigma_{or}^{abs}$ (from work <sup>17</sup> ) | 7 - $\sigma_{or}^{abs}$ (from work <sup>18</sup> ) |
| 4 - $\sigma_{or}^{abs}$ (from work <sup>20</sup> ) |  |

Up to now, it is possible to compare the results obtained in this work only for chemo-ionization cross sections since all previous experiments were carried out with unpolarized particles. As seen from Figure 4, the chemo-ionization cross section obtained in this work is in good agreement with the literature data (considering that the accuracy of the experiments declared in the cited works is about 10-20%). In Figure 4 the results of our previous work<sup>12</sup> (for chemo-ionization cross section) are also presented. As follows from Figure 3 and Table 1, the cross sections calculated in<sup>12</sup> were significantly underestimated. This is because, as mentioned above, the calculation in<sup>12</sup> was carried out for internuclear distances of 3-15  $a_0$  and at the collisional energies from 2·10 to 4 - 6·10<sup>-3</sup> a.u. Such limitations led to a significant underestimation of the studied cross-sections

## 5. Conclusions

As follows from the paper, the polarization transfer in the collision of polarized metastable helium atoms is due to the simultaneous processes of spin exchange and chemo-ionization. These two processes influence each other.

The spin-exchange cross sections in the case of a collision of two metastable helium atoms differ significantly from the point when the chemo-ionization does not accompany the spin-exchange process (for example, in the case of a collision of two alkali atoms in the ground state). The simultaneous occurrence of elastic and inelastic processes leads to the spin exchange cross-section depending on the inelastic process. The effect of chemo-ionization on

the spin exchange cross section is due to the imaginary part of the scattering phases on singlet and triplet terms in expressions for spin exchange cross sections.

## References

- 1- W. Happer, Optical pumping. Rev. Mod.Phys., **1972**,169.
- 2- R.C. Saptarshi, S. Pradhan, Optical pumping and relaxation of atomic population in assorted conditions, J. Phys. B: At. Mol. Opt. Phys., **2022**, 55, 165502.
- 3- M. Pawlak, P. Żuchowski, P. Jankowski, Kinetic isotope effect in low-energy collisions between hydrogen isotopologues and metastable helium atoms: Theoretical calculations including the vibrational excitation of the molecule, J. Chem. Theory Comput., **2021**, 17, 1008.
- 4- T.J. Beams, I.B. Whittingham, G. Peach, Autoionization of spin-polarized metastable helium in tight anisotropic harmonic traps, Phys. Rev., **2007**, A 76, 062707.
- 5- R.A. Zhitnikov, V.A. Kartoshkin., Metastability exchange in a mixture of He<sup>3</sup> and He<sup>4</sup> isotopes, Sov.Phys. JETP., **1976**, 44, 924-930.
- 6- D. Vranceanu, H.R. Sadeghpour, Spin polarization transfer in ground and metastable helium atom collisions, New Journal of Physics, **2010**, 12, 065039.
- 7- P. Nikolaou, B.M. Goodson, E.Y. Chekmenev, NMR Hyperpolarization Techniques for Biomedicine, Chem. Eur. J., **2015**, 21, 3156.
- 8 - R.J.W. Stas, J. M. McNamara, W. Hogervorst, W. Vassen, Homonuclear ionizing collisions of laser-cooled metastable helium atoms, Phys. Rev. A, **2006**, 73, 032713.
- 9 - M.J. Namara, R.J. W. Stas, W. Hogervorst,

- W. Vassen, Heteronuclear ionizing collisions between laser-cooled metastable helium atoms, *Physical Review A*, **2007**, 75, 062715.
- 10- V.A. Kartoshkin, Chemoionization and spin exchange in collisions of excited metastable helium atoms with sodium atoms in the ground state: II. The calculation of cross sections for chemo-ionization and spin exchange, *Optics and Spectroscopy*, **1998**, 85, 177.
- 11- P.J. Leo, V. Venturi, I.B. Whittingham, J.F. Babb, Ultracold collisions of metastable helium atoms, *Phys. Rev. A*, **2001**, 64, 042710.
- 12- N.F. Mott, H.S.W. Massey, *The Theory of Atomic Collisions*, 3rd ed., Oxford: Clarendon Press, **1965**, 858.
- 13- E. Wigner, Ueber die Erhaltungssätze in der Quantenmechanik, *Gotting.Nachr.*, **1927**, 4, 375-381.
- 14- A. Dalgarno, M.J. Rudge, Spin-Change Cross-Sections for Collisions between Alkali Atoms, *Proc. Roy. Soc.*, **1965**, A286, 519.
- 15- V.D. Melnikov, V.A. Kartoshkin, G.V. Klementiev, Spin exchange, accompanying with the nonelastic process under the collision of two atomic particles with  $S_1=S_2=1$  electron spins, *ZhurnalTekhnicheskoiFiziki*, **1988**, 58, 830.
- 16- M.W. Müller, A. Merz, W. RufM, H. Hotop, W. Meyer, M. Movre, Experimental and theoretical studies of the Bi-excited collision systems  $\text{He}^*(2^3\text{S}) + \text{He}^*(2^3\text{S}, 2^1\text{S})$  at thermal and subthermal kinetic energies, *Z. Phys. D.*, **1991**, 21, 89-112.
- 17- V.A. Kartoshkin, Chemoionization and spin exchange in collisions of triplet metastable atoms in a mixture of helium isotopes, *Optics and Spectroscopy*, **2008**, 105, 657-662.
- 18- R. Deloche, P. Monchicourt, High-pressure helium afterglow at room temperature, *Phys. Rev.*, **1976**, A 13, 1140.
- 19- B.J. Garrison, W.H. Miller, H.F. Shaefer, Penning and associative ionization of triplet metastable helium atoms, *J. Chem. Phys.*, **1973**, 59, 3193.
- 20- G.D. Magnuson, S.Y. Tang, Chemo-ionization in collisions of metastable helium with metastable helium, *J. Chem. Phys.*, **1978**, 68, 5112.
- 21- P.L. Bender, Interpretation of Frequency Shifts Due to Electron Exchange Collisions, *Phys. Rev.*, **1964**, A134, A1174.
- 22- M. Baranger, Spectral line broadening in plasmas, *Pure and Applied Physics*, **1962**, 13, 493-548.

## Appendix 1

Consider the interaction of two metastable helium atoms in the  $2^3S_1$  state. Each of these atoms has an electron spin  $S=1$ . Helium atoms are diamagnetic in the ground state ( $1^1S_0$ ) and paramagnetic in the metastable state ( $2^3S_1$ ). As noted above, the interaction between elastic metastable helium atoms (spin exchange) and inelastic (chemo-ionization) processes occurs simultaneously. These processes

$$P = \begin{pmatrix} P_{11} & 0 & 0 \\ 0 & P_{22} & 0 \\ 0 & 0 & P_{33} \end{pmatrix} \quad \rho = \begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix} \quad (\text{A-1})$$

We consider the situation of the interaction of a spin polarized metastable helium atom A (only diagonal elements are present in the density matrix) with an unpolarized helium atom B.

In neglecting the correlation between the P and  $\rho$  matrices, the interacting particles comprise a system that can be described by the density matrix  $\Phi$  as

influence observables in the experiment quantities – orientations and alignments.

Begin our consideration with the spin exchange process. Since the electronic spin of the helium atom A is  $S_1=1$ , and the electronic spin of the helium atom B is  $S_2=1$  too, the particles under study can be described by the corresponding density matrices P (particle A) and  $\rho$  (particle B) of  $3 \times 3$  and  $3 \times 3$  dimensionality. In this case, the P and  $\rho$  matrices have the form

$$\Phi = \rho \otimes P \quad (\text{A-2})$$

where the sign  $\otimes$  denotes the direct matrix product.

The transformation 21 gives the transition to the electron-spin-coupled representation

$$\Phi^c = \Gamma \Phi \Gamma^c \quad (\text{A-3})$$

$m_1, m_2$ $S, M_S$	1,1	0,1	-1,1	1,0	0,0	-1,0	1,-1	0,-1	-1,-1
2,2	1								
2,1		$1/\sqrt{2}$		$1/\sqrt{2}$					
2,0			$1/\sqrt{6}$		$\sqrt{2}/\sqrt{3}$		$1/\sqrt{6}$		
2,-1						$1/\sqrt{2}$		$1/\sqrt{2}$	
2,-2									1
1,1		$-1/\sqrt{2}$		$1/\sqrt{2}$					
1,0			$-1/\sqrt{2}$				$1/\sqrt{2}$		
1,-1						$-1/\sqrt{2}$		$1/\sqrt{2}$	
0,0			$1/\sqrt{3}$		$-1/\sqrt{3}$		$1/\sqrt{3}$		

Here  $S_1=1$  and  $S_2=1$  are the electron spins of the metastable helium atoms, and  $m_1$  and  $m_2$  are the projections of spins. The spins  $S_1$  and  $S_2$  give the total spin  $S$  with the values 0, 1 or 2 and the magnetic

quantum number  $M_S = m_1 + m_2$ , which corresponds to the molecular wave function (See Eq.(7) and Eq(8)).

The evolution of the density matrix  $\Phi^c$  in the spin exchange occurs according to (A-4) <sup>21,22</sup>,

$$\left( \frac{d\Phi_{pq}^c}{dt} \right)_{coll} = -(w - id)\Phi_{pq}^c (1 - \delta_{pq}) \quad (\text{A-4})$$

Where  $w = -\text{Im } H_{pq}$ ,  $d = \text{Re } H_{pq}$ ,  $\delta_{pq}$  is the Kronecker symbol.

$$\left( \frac{d\Phi}{dt} \right)_{coll} = \Gamma^c \left( \frac{d\Phi^c}{dt} \right)_{coll} \Gamma \quad (\text{A-5})$$

To get kinetics equations describing the evolution of the density matrices P and  $\rho$  we need to compute the

The return to the uncoupled representation of moments  $\Phi$  is carried out using the transformation

trace of the Eq.(A-5), for example,

$$\frac{d\rho}{dt} = \text{Tr}_{B_e} \left[ \left( \frac{d\Phi}{dt} \right)_{coll} \right] \quad (\text{A-6})$$

Using the relations (A-2)-(A-6), we can write out the expression for the longitudinal orientation of particle A and B.

In this case, the values of longitudinal orientations and alignment can be represented in the following form:

here  $\langle S_A \rangle_z$  is the longitudinal orientation of particle

A [ $\langle S_A \rangle^z = (\rho_{11} - \rho_{33})$ ],  $\langle S_B \rangle^z$  is the longitudinal orientation of particle B [ $\langle S_B \rangle^z = (P_{11} - P_{33})$ ],

$\langle Q_B \rangle^{zz}$  is the longitudinal components of the electronic alignment of the particle

B [ $\langle Q_B \rangle^{zz} = \frac{1}{2}(P_{11} + P_{33}) - P_{22}$ ]. Here  $P_{ii}$  and  $\rho_{ii}$  are

the diagonal matrix elements of the density matrices  $P$  and  $\rho$ , describing atoms A and B.

Based on the above, it is possible to construct kinetic equations that describe the evolution of the quantities observed in the experiment - orientation and

alignment. The results are presented by expressions (19) and (20).

Consider expressions (19) and (20). Square brackets in expressions (19) and (20) characterize the change in the orientation of atom A as a result of its collision with atom B. In this case, the sign (-) before the bracket indicates the loss of orientation of the ensemble of atoms A during the collision. The sign (+) before the bracket corresponds to the acquisition of orientation by the atom A. Parentheses, which include sections with the index tr, are associated with the spin exchange process. The chemo-ionization process determines those that have sections with the index abs. The last two square brackets in (19) and (20) are connected by the influence of alignment  $\langle Q \rangle$  on the change in the orientation of A atoms in the processes of spin exchange and chemo-ionization. Since the value of orientation and alignment in experiments on the optical orientation of atoms, as usual, is not large ( $\leq 10\%$ ), the terms with the product  $\langle S \rangle \langle Q \rangle$  can be neglected.