Low energy collisions of spin-polarized metastable argon atoms with ground state argon atoms

Taillandier-Loize, T.; Perales, F.; Baudon, J.; Hamamda, M.; Bocvarski, V.; Ducloy, Martial; Correia, F.; Fabre, N.; Dutier, G.

2018

Taillandier-Loize, T., Perales, F., Baudon, J., Hamamda, M., Bocvarski, V., Ducloy, M., . . . Dutier, G. (2018). Low energy collisions of spin-polarized metastable argon atoms with ground state argon atoms. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 51(8), 085202-. doi:10.1088/1361-6455/aab52a

<https://hdl.handle.net/10356/87220>

<https://doi.org/10.1088/1361-6455/aab52a>

© 2018 IOP Publishing Ltd. All rights reserved. This is an author-created, un-copyedited version of an article accepted for publication in *Journal of Physics B: Atomic, Molecular and Optical Physics*. IOP Publishing Ltd is not responsible for any errors or omissions in this version of the manuscript or any version derived from it. The definitive publisher authenticated version is available online at <https://doi.org/10.1088/1361-6455/aab52a>.

Downloaded on 26 Apr 2023 17:47:52 SGT

Low energy collisions of spin-polarized metastable Argon atoms with ground state Argon atoms

T. Taillandier-Loize^{1,2}, F. Perales^{1,a}, J. Baudon¹, M. Hamamda¹,
V. Bocvarski^{3#}, M. Ducloy^{1,4}, F. Correia¹, N. Fabre¹ and G. Dutier¹

¹Laboratoire de Physique des Lasers (UMR-CNRS 7538), Université Paris 13, 99 Av. J.B. Clément, 93430-Villetaneuse, France

²Télécom SudParis, Université Paris-Saclay, 9 rue Charles Fourier- 91 011 Evry Cedex, France

³Institute of Physics - Belgrade, Pregrevica 118, 11080-Zemun, Serbia

⁴PAP, School of Physical and Mathematical Sciences, Nanyang Technological University, 637371 Singapore

#Deceased

^a Corresponding author: francisco.perales@univ-paris13.fr

Abstract

The collision between a spin polarized metastable argon atom in $\text{Ar}^* (3p^5 4s, ^3P_2, M = +2)$ state slightly decelerated by the Zeeman slower-laser technique and a co-propagating thermal ground state argon atom $\text{Ar} (3p^6, ^1S_0)$, both merged from the same supersonic beam, but coming through adjacent slots of a rotating disk, is investigated at center-of-mass energies ranging from 1 to 10 meV. The duration of the laser pulse synchronised with the disk allows the tuning of the relative velocity and thus the collision energy. At these sub-thermal energies, the “resonant metastability transfer” signal is too small to be evidenced. The energy explored range requires using indiscernibility amplitudes for identical isotopes to have a correct interpretation of the experimental results. Nevertheless, excitation transfers are expected to increase significantly at much lower energies as suggested by previous theoretical predictions of potentials $2g (^3P_2)$ and $2u (^3P_2)$. Limits at ultra-low collisional energies of the order of 1 mK (0.086 μeV) or less, where gigantic elastic cross-sections are expected, will be also discussed. The experimental method is versatile and could be applied using different isotopes of Argon like ^{36}Ar combined with ^{40}Ar , as well as other rare gases among which Krypton should be of great interest thanks to the available numerous isotopes present in a natural gas mixture.

Introduction

Merged-beams technique is an experimental method developed in the mid-1960's to measure in absolute units total charge transfer cross sections at low energies [1-2]. The thematic is rich and concerns not only ion-atom or molecule and electron-ion or molecular ion, but also atom-atom or molecule and more generally reactivity, behaviour of plasmas, discharges, etc. For a review see reference [3] and references there in. Symmetric systems are of particular importance in the case of cold trapped atoms, especially with rare gases [4]. Usually rate equations are used to describe such situation. However to properly take in account the symmetry problem, the diffusion amplitudes and their symmetry need to be precisely described, as it will be the case in the present paper. Actually experiments cannot go usually to a low enough energy where indistinguishability of nuclei (or not, when different isotopes are involved), has to be taken into account. In this article, we will show that the concept of indistinguishability (or indiscernibility) is needed to properly study, theoretically and experimentally, the low energy collisions, when an on axis-polarized $M = +2$ $\text{Ar}^* (3p^5 4s, ^3P_2)$ pulsed beam is catch up by a ground state $\text{Ar} (3p^6, ^1S_0)$ pulsed beam. By the past, considerable theoretical effort has been devoted to resonant charge exchange of protons with hydrogen and deuterium atoms [5-8]. An interesting fact is that towards low collision energies, charge transfer cross sections increase only when the charge is carried by the heavier atom [6]. Charge transfer or exchange involves r^{-4} long range attractive potentials where solutions of the radial Schrödinger equation can be obtained through Mathieu functions. Excitation or metastability transfer is governed by the attractive dipole-dipole long range r^{-6} potentials connecting to wells or repulsive humps at intermediate or short distances. As an example of such problems, it was observed in [9], time of flight spectra showing metastability

transfers in helium between the singlet state $\text{He}^*(1s2s, {}^1S_1)$, the triplet $\text{He}^*(1s2s, {}^3S_1)$ and the ground state $\text{He}(1s^2, {}^1S_0)$. This was done in the same Helium 4 supersonic beam after electronic excitation and was proved coming from a secondary process between the metastable and the superabundant ground state. The $\text{He}(1 {}^1S) - \text{He}^*(2 {}^3S)$ system has also been studied theoretically in [10] where an elastic cross section of about $5000 \text{ \AA}^2$ is predicted at ultra-low energy through the s-wave scattering, in which the only $c^3\Sigma_g^+$ potential is involved. Recently, an experiment has been done merging two optical traps, one for $\text{Ar}^*(3p^5 4s, {}^3P_2)$ and one for ground state $\text{Ar}(3p^6, {}^1S_0)$, thus evidencing a huge elastic cross section estimated to be around $600\,000 \text{ \AA}^2$ in the mK temperature range [11].

The present article is first dedicated to the theoretical aspect of the problem and describes the realistic gerade and ungerade analytic potentials we used. Are only relevant for our problem (in the range $R > 6 a_0$), the so-called $\Omega = 2_g$ and 2_u potentials of the $\text{Ar}^*(3p^5 4s, {}^3P_2) - \text{Ar}(3p^6, {}^1S_0)$ system. They reasonably agree with data points up to $R = 12 a_0$, based on *ab initio*, configuration-interaction (C.I.) calculations from Spiegelmann et al [12, 13]. The long range behaviour in C_6/R^6 taken from Beijerinck et al [14] has been chosen. Then, phase-shifts, generated by the JWKB approximation are calculated. Their validity in the energy range 1 to 10 meV is discussed. We also describe wave-functions used to solve the radial Schrödinger equation. Scattering amplitudes with their related symmetries are defined in the center-of-mass (CM) reference frame. This part first presents the terminology and then the calculation of the different differential and total cross sections. Their validity and relevance are discussed in what concerns their confrontation to describe measurements. Also, the ultra-low energy limit is discussed, presenting a large scattering length, and a probable large number of bounded states for the $\text{Ar}^*({}^3P_0 \text{ and } {}^3P_2) + \text{Ar}$ system. At the end of this section, we shall explain why exchange of velocities from two different sources can be very efficient at much lower energy, justifying the results of [15] where final metastable state populations of 3P_0 and 3P_2 are in the proportion 33.8% to 66.2% rather than the usual stoichiometric proportion 1/6 to 5/6 obtained after electronic collisions.

In its principle the present experiment is close to that described in a previous paper [16] in which the exchange process in argon has been investigated at larger CM energies (8 meV – 27 meV). Owing to improvements described here after, lower CM energies have become accessible. The present experiment is presented in detail in the second part of the paper. Then, few time of flight spectra are presented and analysed. From these data it has been possible to derive 2 total cross sections for indistinguishable partners at 1 and 2 meV and partial cross sections to about 10 meV. Velocity exchange signals are not visible as peaks in our time of flight spectra because their population is still too small and we do not have a resolution high enough (counting statistics) to distinguish them from the incident metastable flux arriving at the same time.

The conclusion will summarize the strong points developed through the article. A possible extension of the present experimental method to Argon isotope 36 and to Krypton, which possesses a rather large number of significantly populated isotopes, including one Kr 83 with a 9/2 nuclear spin would be interesting.

Part I-Theoretical aspect

I-1 Potential Energies and Phase shifts determination

Our interpretation will be restricted to the unique case of argon, for which we introduce new analytical expressions of 2_g and 2_u potentials as close as possible to the available *ab initio* potential data points of [12, 13]:

$$V_{(g,u)}(R) = A_{(g,u)} R^{\beta_{(g,u)}} e^{-\alpha_{(g,u)} R} - \left[\frac{C_6}{R^6} + \frac{C_8}{R^8} + \frac{C_{10}}{R^{10}} \right] f_c(R) \quad (1)$$

where A , β , α are fitting parameters and $f_c(R) = \Theta(R - R_c) + \Theta(R_c - R) \times e^{-\left(\frac{R_c}{R} - 1\right)^2}$. The cut-off function f_c at $R_c = 21.98 a_0$ is defined as in [19] where Θ is the unit step function. Note that the leading long range van der Waals coefficient C_6 is shared by gerade and ungerade potentials. This fact could be justified by the similar electronic structure of our system as in $\text{He}(2^3\text{S})\text{-He}(2^3\text{P})$ in [18], the dipole-dipole dispersion van der Waals coefficient, is taken from reference [14], C_8 and C_{10} are fitting parameters since to our knowledge dipole-quadrupole and quadrupole-quadrupole dispersion coefficients are unfortunately not available in the literature. They should be different for u and g potentials, but their contribution is expected to be very small. Their values in atomic units are listed below in table I.

At shorter distance range, these potentials present a steep barrier, which leads to a deep well at intermediate inter-nuclear distance ($R \sim 10.5 a_0$). The well depth $\varepsilon \sim 9 \text{ meV}$ is comparable to the collision energies we are exploring here in our experiment (see figure 1). Values of parameters are given in atomic units and are listed in table I.

	A	β	α
V_g	40.8831	1.47175	2.09
V_u	4.3981	3.44355	2.14084
V_g and V_u	C_6	C_8	C_{10}
	328.57	264004	-18451910

Table I Parameters in atomic units involved in potential energies $2g$ and $2u$ of formula (1).

Figure 1. Filled and open circles are respectively gerade and ungerade data points from [11,12], continuous and dashed lines are our analytical expressions (validity is ensured for $R > 6 a_0$). Notice that points at $R = 12 a_0$ are already very close to each other.

The validity of JWKB approximation has been checked by evaluating the phase shifts numerically from the radial equation [17]. The JWKB phase-shifts, computed using a Gauss

sum rule [18], originate from the relative phase between on and off potentials, from the turning points to infinite interatomic distance:

$$\delta_{(g,u)}^l(k) = \int_{r_{0(g,u)}^l}^{\infty} \sqrt{k^2 - U_{(g,u)} - \frac{\left(l + \frac{1}{2}\right)^2}{r^2}} dr - \int_{\frac{l+\frac{1}{2}}{k}}^{\infty} \sqrt{k^2 - \frac{\left(l + \frac{1}{2}\right)^2}{r^2}} dr \quad (2)$$

Their expressions reduce as follows:

$$\delta_{(g,u)}^l(k) = \int_{r_{0(g,u)}^l}^{\infty} \sqrt{k^2 - U_{(g,u)} - \frac{\left(l + \frac{1}{2}\right)^2}{r^2}} dr - kr_{0(g,u)}^l + \frac{\left(l + \frac{1}{2}\right)\pi}{2} \quad (3)$$

Terms $U_{(g,u)}(R) = 2\mu \frac{V_{(g,u)}(R)}{\hbar^2}$ are reduced potentials where $\mu = m/2$ is the reduced mass of two ^{40}Ar atoms, k^2 is the reduced collision energy, and $r_{0(g,u)}^l$ are distances of closest

approach, *i.e.* the largest roots of equations $k^2 - U_{(g,u)}(r) - \frac{\left(l + \frac{1}{2}\right)^2}{r^2} = 0$.

I-2 Wave function, scattering amplitudes and cross sections

Let us consider the initial merging collision partners where a and b stand for recognizable nuclei although identical in our case, the sign $*$ stands for the metastable character, here the electronic excitation and V_{slow} , V_{fast} are the respective velocities of a and b in the laboratory frame. Quantization axis is the Zeeman slower axis, *i.e.* the direction of the magnetic field, necessary for the laser to be resonant during the slowing, parallel to both beams directed towards the detector. The collision is described as follow:

The wave function depends on electronic coordinates referring to atoms a and b that only separates at large molecular distance R .

$$\Psi(\vec{r}_a, \vec{r}_b, \vec{R}) = \varphi(\vec{R}) \Phi(\vec{r}_a, \vec{r}_b, R) \rightarrow \varphi(\vec{R}) \Phi(\vec{r}_a) \Phi(\vec{r}_b) \text{ at } R \rightarrow \infty$$

The gerade-ungerade symmetry makes that the proper basis of states is the one where we do not know to which nucleus belongs the electronic excitation (represented by a star $*$):

$$\Phi_g = \frac{\Phi_a^* \Phi_b + \Phi_a \Phi_b^*}{\sqrt{2}}, \Phi_u = \frac{\Phi_a^* \Phi_b - \Phi_a \Phi_b^*}{\sqrt{2}} \quad (4)$$

The incoming state projected over the molecular axis is: $\varphi(\vec{R}) \Phi_a^* \Phi_b = \frac{\varphi(\vec{R})(\Phi_g + \Phi_u)}{\sqrt{2}}$.
Then the outgoing state writes at large R :

$$\varphi(\vec{R})\Phi(\vec{r}_a, \vec{r}_b, R) = e^{ikz} \frac{\Phi_g + \Phi_u}{\sqrt{2}} + \frac{e^{ikR}}{R} \left[\frac{f_g(\theta)\Phi_g + f_u(\theta)\Phi_u}{\sqrt{2}} \right] \quad (5)$$

In the center of mass system (see figure 2), the molecular coordinate along the incident direction is $z = R \cos \theta$ and the scattering amplitudes for the gerade and ungerade potentials are defined as:

$$f_{(g,u)}(\theta) = \frac{1}{2ik} \sum_{l=0}^{\infty} (2l+1) (e^{2i\delta_{(g,u)}^l} - 1) P_l(\cos \theta) \quad (6)$$

In a more usual way (5) is written as follow:

$$\varphi(\vec{R})\Phi(\vec{r}_a, \vec{r}_b, R) = e^{ikz} \frac{\Phi_g + \Phi_u}{\sqrt{2}} + \frac{e^{ikR}}{R} \left[\left(\frac{f_g(\theta) + f_u(\theta)}{2} \right) \frac{\Phi_g + \Phi_u}{\sqrt{2}} + \left(\frac{f_g(\theta) - f_u(\theta)}{2} \right) \frac{\Phi_g - \Phi_u}{\sqrt{2}} \right]$$

i.e:

$$\varphi(\vec{R})\Phi(\vec{r}_a, \vec{r}_b, R) = e^{ikz} \Phi_a^* \Phi_b + \frac{e^{ikR}}{R} \left[\left(\frac{f_g(\theta) + f_u(\theta)}{2} \right) \Phi_a^* \Phi_b + \left(\frac{f_g(\theta) - f_u(\theta)}{2} \right) \Phi_a \Phi_b^* \right] \quad (7)$$

, which gives more physical insight since the wave function is projected on the detection basis set and shows clearly what is commonly called a “direct” amplitude and an “exchange” amplitude.

Figure 2. Kinematics of the collision in laboratory and center of mass reference frames for a fast ground state beam colliding with a slowed metastable beam at an angle β . θ is the polar CM angle and $\phi = 0$, $\phi = \pi$ the in-plane azimuthal disks of metastable detected direction R .

Note that, *a priori*, in the case of a collision with a spin polarized atom $M = +2$, scattering amplitudes should allow transitions to all M states referred to the Zeeman-laser axis called also sometimes “excitation frame”. This is equivalent to consider transitions quantized in the molecular frame (Ω being the projection of angular momentum on the molecular axis R),

from $\Omega = 2$ towards $\Omega = 1$ and $\Omega = 0$ and not only to $\Omega = 2$ as assumed in our approach. Several reasons are favourable to simplify and reduce the problem to a $2 - 2$ process: (i) quantization direction, incident direction and detection direction are merged when $\beta = 0$, β being the angle between lab-velocities (fig.2) to a good approximation, thus the only initial magnetic quantum numbers M or $\Omega = 2$ can be considered, (ii) $\Omega = 2 - 1$ and $\Omega = 2 - 0$ transitions exist but, because of the rather small effect of rotational coupling at low energy, they are expected to be smaller than $\Omega = 2 - 2$ transitions. Under this condition we do not have to consider any $\varphi - \varphi + \pi$ azimuthal dependence in the scattering amplitudes. In a full approach, for example when spin transitions in the laboratory frame $M = 2 - 1$ have to be considered, the scattering amplitude has an imaginary exponential term in $\Delta M \varphi$ appearing in spherical harmonics functions, where ΔM is the difference of M -s, *i.e.* $\Delta M = 1$ that makes it φ dependant (see for example [21]). In our case however, to a good approximation that we will check *a posteriori*, only two $\Omega = 2$ - molecular states have been considered, namely 2_g and 2_u and scattering amplitudes will be considered with azimuthal isotropy around the main incident beam direction z in the CM frame (see figure 2).

For identical nuclei which is the case for Argon 40 to better than 99.5 % (isotope 39.962384 from the NIST tables), we have to consider the inversion operator I exchanging indistinguishable nuclei. Operator I transforms $\vec{R}(R, \theta, \varphi)$ into $-\vec{R}(R, \pi - \theta, \varphi + \pi)$ and commutes with the total Hamiltonian, thus adding a new part in the wave function expressing that nucleus (a) and nucleus (b) cannot be distinguished in the molecular wave function. As a consequence direction (θ, φ) and direction $(\pi - \theta, \varphi + \pi)$ have to be taken into account for interference of direct and exchange amplitudes that would be detected by a conceptual center of mass frame detector (see figure 2). Thus, the total asymptotic wave function replacing (5) is now:

$$\Psi(\vec{r}_a, \vec{r}_b, R) = \frac{\varphi(\vec{R})\Phi(\vec{r}_a, \vec{r}_b, R) + \varphi(-\vec{R})\Phi(\vec{r}_a, \vec{r}_b, R)}{\sqrt{2}}. \quad (8)$$

At large R , it can be equivalently written:

$$\frac{1}{\sqrt{2}} \left[e^{ikz} \frac{\Phi_g + \Phi_u}{\sqrt{2}} + e^{-ikz} \frac{\Phi_g - \Phi_u}{\sqrt{2}} \right] + \frac{1}{\sqrt{2}} \frac{e^{ikR}}{R} \left[\frac{[f_g(\theta)\Phi]_g + f_u(\theta)\Phi_u + f_g(\pi - \theta)\Phi_g - f_u(\pi - \theta)\Phi_u}{\sqrt{2}} \right]$$

. The outgoing spherical wave in the second term can be expressed in the proper basis as:

$$\frac{e^{ikR}}{R} \left(\frac{f_g(\theta) + f_u(\theta) + f_g(\pi - \theta) - f_u(\pi - \theta)}{2} \right) \frac{\Phi_g + \Phi_u}{\sqrt{2}} + \frac{e^{ikR}}{R} \left(\frac{f_g(\theta) - f_u(\theta) + f_g(\pi - \theta) + f_u(\pi - \theta)}{2} \right) \frac{\Phi_g - \Phi_u}{\sqrt{2}}$$

or equivalently:

$$\frac{e^{ikR}}{R} \left(\frac{f_g(\theta) + f_u(\theta) + f_g(\pi - \theta) - f_u(\pi - \theta)}{2} \right) \Phi_a^* \Phi_b + \frac{e^{ikR}}{R} \left(\frac{f_g(\theta) - f_u(\theta) + f_g(\pi - \theta) + f_u(\pi - \theta)}{2} \right) \Phi_a \Phi_b^*$$

(9), now showing the full role of interferences between direct and exchange amplitudes in respectively the direction (θ, φ) and the opposite direction $(\pi - \theta, \varphi + \pi)$.

As a general fact, all processes for the elastic collisional system $\text{Ar}^*(3p^5 4s, ^3P_2) - \text{Ar} (3p^6, ^1S_0)$ are detected with the indiscernible amplitude:

$$f_{ind}(\theta) = \frac{1}{2} [f_g(\theta) + f_u(\theta) + f_g(\pi - \theta) - f_u(\pi - \theta)] \quad (10)$$

Let us call as it is commonly done - “direct” and “exchange” amplitudes the following combined gerade-ungerade amplitudes:

$$f_{dir}(\theta) = \frac{1}{2} [f_g(\theta) + f_u(\theta)] \quad \text{and} \quad f_{exc}(\theta) = \frac{1}{2} [f_g(\theta) - f_u(\theta)] \quad (11)$$

Let us imagine that we have two CM detectors like in figure 2. Then we shall detect metastable atoms in the region $\theta \approx 0$, $\varphi = 0$ at velocity V_{slow} with the indiscernible amplitude,

$f_{ind}^{slow}(\theta \approx 0, \varphi = 0) = f_{dir}(\theta) + f_{exc}(\pi - \theta)$ and V_{fast} in the opposite direction around $\theta' \approx \pi, \varphi = \pi$, with the indiscernible amplitude $f_{ind}^{fast}(\theta' \approx \pi, \varphi = \pi) = f_{dir}(\pi - \theta) + f_{exc}(\theta)$. Let's assume that the angle β between the two beams is different from 0, for example $\pi/2$. It is tempting to say that V_{fast} atoms measured in the laboratory frame, without any spatial noise in the signal, are the signature of a metastability transfer. AS WELL, if we assume two merging beams, such that $\beta=0$, one could think of measuring in a time of flight spectrum the fast atoms by their earlier arrival time and thus, conclude in the same way as before. This is a too naive image in both cases as we will see later in our particular range of energies. Now the differential cross section (DCS) as a function of θ is:

$$\frac{d\sigma}{d\theta}(\theta) = \left[\frac{1}{2} [f_g(\theta) + f_u(\theta) + f_g(\pi - \theta) - f_u(\pi - \theta)] \right]^2. \quad (12)$$

We have computed this DCS(θ) for several energies. At CM energy of 65 meV indiscernibility differential cross section at $\theta \sim \pi$ is well reproduced by the exchange differential cross section (see figure 3): $\frac{d\sigma}{d\theta}(\theta \sim \pi) \approx \left[\frac{1}{2} [f_g(\theta) - f_u(\theta)] \right]^2$.

It looks like “direct” and “exchange” amplitudes could give a quasi-correct interpretation of the resonant metastability transfer at 65 meV.

In the past, resonant metastability exchange between identical atoms, of the type $X^*(^3P_2) + X(^1S_0) \rightarrow X + X^*$ where $X = \text{Ar}$ or other rare gases, has been studied experimentally and theoretically in a 90° crossed beam apparatus at thermal energy using time of flight techniques, *i.e.* $E_{th} = 63.5$ meV corresponding to 559 m/s and also at hyperthermal energies. The exchange phenomenon is measured by the detector facing the initial ground state beam, thus being sensitive to the 90° laboratory angle or equivalently at the peak of maximum metastability transfer. In this domain of energy, where indiscernibility of the nuclei plays a little role, the theory is successful, only taking into account the gerade-ungerade symmetry of the X_2^* molecular system and computing the differential cross section with a so-called exchange diffusion amplitude, *i.e.* one half of the difference between gerade and ungerade amplitudes. Evidently, this signal is maximum at an angle of 90° in the laboratory, corresponding to the maximum polar angle $\theta = \pi$, in the center of mass referential frame where the exchange amplitude has to be evaluated [22].

On another hand, at 5 meV, the exchange differential cross section is well under the indiscernibility differential cross section (see figure 3). Consequently the exchange of velocity at large angle cannot be clearly defined. Indeed at large CM angle, direct and exchange amplitudes combine. This happens in a quantum way as interference of these two possibilities cannot be broken by the measurement in an experiment.

Figure 3. Modified differential cross sections ($2\pi \sin\theta \times \frac{d\sigma}{d\theta}$) in a_0^2 as function of CM angle θ for : direct (full line), exchange (dashed line), and in case of indiscernibility (dotted line) processes, at thermal energy (65 meV), low energy (5 meV) and very low energy (0.2 meV).

Notice than the reverse proposition is not true, *i.e.* the direct amplitude is never negligible as a part of the indiscernible amplitude, in the small angle domain. In this sense at low energies, the exchange concept can be lost but we can still speak of a majority of “direct” events at small angles. This is not guaranteed at lower energy that we cannot measure here but rather calculate. As an example, it is seen that at $E=0.2$ meV in figure 3, the exchange concept is indeed brought back.

From the usual expression of gerade and ungerade scattering amplitudes, we get the total gerade and ungerade cross section by integration over the full center of mass solid angle:

$$\sigma_{(g,u)} = \int_0^\pi 2\pi \sin\theta |f_{(g,u)}(\theta)|^2 d\theta, \text{ leading to the well-known simple forms:}$$

$$\sigma_{(g,u)} = \frac{4\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \sin^2 \delta_{(g,u)}^l \quad (13)$$

and for the total cross section, $\sigma_T = \frac{1}{2}(\sigma_g + \sigma_u)$. Note that all these cross sections can be

expressed with their tangent form $\sigma_{(g,u)} = \frac{4\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \frac{\tan^2 \delta_{(g,u)}^l}{1 + \tan^2 \delta_{(g,u)}^l}$. Another popular

form using the optical theorem [23] allows us to write: $\sigma_{(g,u)} = \frac{4\pi}{k} \text{Im}(f_{(g,u)}(0))$ which only needs the calculus of the imaginary part of the gerade and ungerade scattering amplitudes at $\theta = 0$. We can also remark that these quantities have not an easy link with measurements,

although it will be proved at the end of this section that σ_g is, in our case, measurable in the s-wave scattering, *i.e.* at ultra-low energy.

When the collisional energy is high enough and if no details in the differential cross section are looked for, or when we use different isotopes (e.g. Argon has a 0.337% ^{36}Ar mass isotope), we are in a simple formalism in which interference between direct and exchange

amplitudes can be neglected. Similarly from $\sigma_{exc} = \int_0^\pi 2\pi \sin\theta |f_{exc}(\theta)|^2 d\theta$ (see figure 4), with $f_{exc}(\theta) = \frac{1}{2}[f_g(\theta) - f_u(\theta)]$, we get the following formula for the so-called exchange cross section [21]:

$$\sigma_{exc} = \frac{\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \sin^2(\delta_g^l - \delta_u^l) \quad (14)$$

Sometimes the equivalent following form is suitable:

$$\sigma_{exc} = \frac{\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \frac{(\tan \delta_g^l - \tan \delta_u^l)^2}{(1 + \tan^2 \delta_g^l)(1 + \tan^2 \delta_u^l)}$$

, for example to include scattering lengths of the gerade and ungerade potentials at ultra-low energy in the s wave scattering, *i.e.* $l = 0$, or in the p wave scattering with $l = 0$ and 1 when different isotopes are used.

Notice also that for a symmetric system, with our potentials beyond $R = 12 a_0$, the difference between gerade and ungerade phase shifts is close to zero and could be computed in the bounded domain from the turning points of $2g$ and $2u$ potentials up to $R=12 a_0$ with:

$$\delta_g^l(k) - \delta_u^l(k) = \int_{r_{bg}^l}^R \sqrt{k^2 - U_g(r) - \frac{(l+\frac{1}{2})^2}{r^2}} dr - \int_{r_{bu}^l}^R \sqrt{k^2 - U_u(r) - \frac{(l+\frac{1}{2})^2}{r^2}} dr$$

However, as we have seen previously, this only holds at energies where σ_{exc} has still a meaning and from a physical point of view the potentials are tested until $R \approx 12 a_0$.

AS WELL from $\sigma_{dir} = \int_0^\pi 2\pi \sin\theta |f_{dir}(\theta)|^2 d\theta$ and $f_{dir}(\theta) = \frac{1}{2}[f_g(\theta) + f_u(\theta)]$ one can prove the following result for the direct cross section: $\sigma_{dir} = \sigma_T - \sigma_{exc}$.

As said before when the collision energy is low enough one has to compute the so-called total

indiscernibility cross section (see figure 4): $\sigma_{ind} = \int_0^\pi 2\pi \sin\theta |f_{ind}(\theta)|^2 d\theta$, with $f_{ind}(\theta) = \frac{1}{2}[f_g(\theta) + f_u(\theta) + f_g(\pi - \theta) - f_u(\pi - \theta)]$. There is no possible further reduction of the formula for σ_{ind} . In our experiment, we will have to integrate the indiscernible DCS over a limited solid angle in the laboratory, this will be reflected by an integration of the indiscernibility DCS at small angles $\theta \approx 0$ for the “direct” events and at large angle $\theta \approx \pi$ for the “exchange” events. In the present collision energy domain, 1-10 meV, the number of “direct” events is very close to the expected direct contribution. Nevertheless at lower energy, direct and exchange amplitudes will interfere (see fig. 3).

We remark that we can write the indiscernible amplitude using new defined gerade and ungerade terms: $f_G(\theta) = \frac{1}{2}[f_g(\theta) + f_g(\pi - \theta)]$ and $f_U(\theta) = \frac{1}{2}[f_u(\theta) - f_u(\pi - \theta)]$, which gives: $f_{ind}(\theta) = f_G(\theta) + f_U(\theta)$.

Using the parity rule of Legendre polynomials $P_l[\cos(\pi - \theta)] = (-1)^l P_l[\cos(\theta)]$, it is easy to prove the following:

$$\begin{aligned} f_g(\theta) &= (2ik)^{-1} \sum_{l=0, l \text{ even}}^{\infty} [(2l+1)(e^{2i\delta_g^l} - 1)] P_l(\cos\theta) \\ f_u(\theta) &= (2ik)^{-1} \sum_{l=1, l \text{ odd}}^{\infty} [(2l+1)(e^{2i\delta_u^l} - 1)] P_l(\cos\theta). \end{aligned} \quad (15)$$

In that way it is possible to compute the indiscernible amplitudes with the only even gerade partial wave phase shift and the odd ungerade partial wave phase shift at all energies.

Notice that at sufficiently low energy ($E < 1$ mK or $0.086 \mu\text{eV}$) the indiscernible amplitude will be expressed with the only $l = 0$ partial wave. Thus, only the gerade amplitude and consequently δ_g^0 is required to infer the upper limit of the total cross section towards the zero energy limit. In other words, at ultra-low energy, amplitudes are isotropic also in θ and reduce to $f_{ind}(\theta) = f_g^0 = (2ik)^{-1} [(e^{2i\delta_g^0} - 1)] = f_g^0$. In conclusion the indiscernibility total cross section has the gerade total cross section as a zero energy limit where the scattering length a

can be defined by: $\sigma_g^0 = \frac{4\pi}{k^2} \sin^2[\delta_g^0(k)] = \frac{4\pi}{k} \text{Im}[f_g^0(k)] = 4\pi a^2$.

According with [16], we get $a = \bar{a} [1 - \tan(\Phi - \pi/8)]$, where $\bar{a}$ is the mean scattering length. We find here a total phase developed by the 2_g potential of $\Phi = 40.64$ rd at energy $E=0$, $\bar{a} = 2^{-3/2} (2 \mu C_6)^{1/4} \Gamma(3/4)/\Gamma(5/4) = 33.50 a_0$. We deduce then $|a| = 55.68 a_0$ and $10\,910 \text{ \AA}^2$ for the 0-energy elastic cross section. Around 12 bound states should exist in the potential well, the energy of the highest one being at -0.00075 meV. It is likely that the details of the true gerade potential strongly contradict these values, this limit being so critical for the 0-energy limit of the gerade cross section. It has also to be noticed that the value $600\,000 \text{ \AA}^2$ obtained for σ_{ind} by merging two dipole traps from reference [10] is not contradictory since their potential is modified by the light shift.

AS WELL, for the p-wave scattering, we have: $f_{ind}(\theta) = f_g^0 + f_u^1 = (2ik)^{-1} [e^{2i\delta_g^0} - 1 + 3(e^{2i\delta_u^1} - 1)\cos\theta]$, which is, this time θ dependent and gives the following total indiscernible cross section:

$$\sigma_{ind} = \frac{8\pi}{k^2} \left(1 - \frac{\cos 2\delta_g^0 + 3\cos 2\delta_u^1}{4} \right) = \frac{4\pi}{k^2} (\sin^2 \delta_g^0 + 3\sin^2 \delta_u^1)$$

If we were considering the system with different isotopes as $^{40}\text{Ar}^* (^3P_2, M=2) + ^{36}\text{Ar} (^1S_0)$

then the total cross section result would be: $\sigma_T = \frac{4\pi}{k^2} (\sin^2 \delta_g^0 + 3\sin^2 \delta_u^1 + \sin^2 \delta_u^0 + 3\sin^2 \delta_g^1)$ and the full exchange cross section (in principle measurable) then reads:

$$\sigma_{exc} = \frac{\pi}{k^2} (\sin^2(\delta_g^0 - \delta_u^0) + 3\sin^2[(\delta_u^1 - \delta_g^1)]) \quad \text{whereas the direct cross section is:}$$

$$\sigma_{dir} = \sigma_T - \sigma_{exc}.$$

By the past it was identified by TOF resolved spectra from [8] that resonant metastability transfer with ground states of Helium 4 occurs continuously in a supersonic expansion submitted to a co-propagating excitation by electron bombardment. This fact was observed for singlet $\text{He}^*(2s \ ^1S_1, \text{internal energy } 20.61 \text{ eV})$ and triplet $\text{He}^*(2s \ ^3S_1, \text{internal energy } 19.82 \text{ eV})$ at sub thermal energies as the nozzle reservoir was cooled to Liquid Nitrogen temperature 77K. The fast peak at early time of flight has a kinetic energy $E_1 = 19.5$ meV and corresponds to metastable atoms being faster at velocity 970 m/s because submitted to an electron impact. This process is called primary process and evolves linearly with pressure in the source chamber. The slow peak at $E_2 = 16.5$ meV (890 m/s) is a resonant metastability transfer and thus comes from a secondary process evolving quadratically with pressure in the source chamber. Collision energy of this process is $\frac{1}{2} (E_1^{1/2} - E_2^{1/2})^2 = 0.062$ meV or equivalently 726

mK. The authors did predict even more efficiency for heavier atoms as neon, argon etc., probably since the recoil being smaller, the lower is the collision energy. In [14] we did the anti-parallel electron bombardment of a thermal argon supersonic beam and then extracted the “exchanged” beam with a 80 μm diameter diaphragm. The recoil, necessary to get an excitation of the atom of 11.5 eV, being estimated to 4.3 m/s, the collision energy is about 0.002 meV or 22.5 mK.

At this collision energy, cross sections involving $^3\text{P}_0$ (see similar potentials 0_g and 0_u in [24]) and $^3\text{P}_2$ have to be already very big and need a proper way, well beyond the JWKB approximation, to be evaluate through the about 10 first partial waves. AS WELL, transfers can be roughly estimated to 30%. We measured a non-stoichiometric ratio of 33.8% $^3\text{P}_0$ and 66.2% $^3\text{P}_2$ by pushing off the peak of magnetic species $^3\text{P}_2$ from the $^3\text{P}_0$ one with a low power resonant laser. We did show then that the metastable beam had a good spatial coherence to diffract even at thermal energy when passing a transmission nano-grating of 100 nm period [25] and in this sense he has the “natural” properties of the ground state nozzle beam.

Figure 4. Left: total indiscernibility cross section and right: total exchange cross section for different isotopes. Cross sections are in atomic units a_0^2 , plotted as functions of CM energies in meV. Notice the 10 factor in cross section scales between the two graphics.

Part II- Experiment

II-1 Description

An argon supersonic beam is partially excited to metastable levels $^3\text{P}_2$ and $^3\text{P}_0$, (ratio 5/6 and 1/6 respectively) by on-axis counter propagating bombardment of electrons (100–150 eV) extracted from an annular oxide cathode (Heat Wave Labs, Inc.). Electron bombardment is more efficient after focussing electrons in an axial magnetic field of about 600 G produced by a coil. Nevertheless the excitation has a low efficiency of about 10^{-7} , and consequently most of the atoms in the beam are at ground state level $3p^6$ ($^1\text{S}_0$). Only metastable atoms, which carry around 11.5 eV of internal energy, are detected. This is easily done with electron multiplier-type detectors like the channeltron and the Delay Line Detector (DLD) (with 2

chevron channel plates) we used (RoentDek Handels GmbH). Time of flight (TOF) and space dependant measurement techniques allow to characterize the thermal velocity $v_i = 553$ m/s of the metastable atomic beam, its full relative dispersion at $1/e$ being $\sim 10\%$ and the full angular aperture or divergence of the beam after the slotted disk being estimated at 3.5 mrad. In order to do a merging collision between slowed 3P_2 and thermal 1S_0 argon atoms we combine two time synchronized techniques: the TOF slotted disk technique to sample the atomic beam (frequency ~ 50 Hz, 3 slots respectively at 0° , 18° and 90°) and the Zeeman slower technique [26]. Usually the Zeeman slower technique is applied continuously in order to fill a magneto-optical trap [27] with an entrance velocity reduced from thermal to a few tens of m/s. The principle of the technique is to slow down the atoms through their interaction at resonance with a counter propagating laser beam. This has to be done compensating the Doppler shift experienced by the decelerated atom. Tuning on resonance of the atomic energy level through the Zeeman effect is realized with a special configuration of longitudinal magnetic field $B(z)$

that can be related linearly to the slowed velocity $v(z)$ by the formula

$$B(z) = \frac{\hbar}{\lambda \mu_B} [v(z) + \lambda \delta_L]$$

. The laser frequency is locked on the $^3P_2-^3D_3$ closed transition at $\lambda = 811.531$ nm and has a negative detuning $\delta_L = -340$ MHz. The circularly polarized ($\sigma+$) laser beam counter propagates with respect to the atom beam after reflection by a golden mirror of diameter $\phi = 27.7$ mm inclined at 45° , and placed at 2.385 m, with a thin rectangular horizontal slit at its center ($15\text{mm} - 150 \mu\text{m}$). This mirror also serves as a secondary emission electrode for the channeltron, whereas a delay line detector is placed well after, at 2.995 m. It is to remark that only 3P_2 atoms can get fully polarized in the $M=+2$ spin sublevel at the entrance of the solenoid where the magnetic field is the highest and subsequently get slowed down. 3P_0 atoms are not affected by the slower and remain at a velocity close to v_i until their detection as fast atoms.

In our experiment, the goal is to slightly decelerate 3P_2 metastable atoms coming from the first slot in order to get caught by fast 1S_0 ground state atoms coming from the next 18° slot. This has to be done before arrival of metastable atoms at the detector position and this fact gives a lower limit to the collision energy, center of mass energy E_{cm} which is in our case, close to 1 meV at the DLD position and 2 meV at the Channeltron position. In this way access to smaller than thermal relative velocities is provided. The third slot at 90° is for calibration purposes and allows eventual corrections for drifts of the metastable beam. To selectively make the atoms unhooked from the slowing process, it is better –and easier – to cut down the laser than turning off the magnetic field. This is done separating by diffraction the laser beam with an acousto-optical modulator (AOM) operated synchronously with the TOF disk for durations of a few ms (see figure 5). Thus a typical sequence for a metastable atom is (i) a free flight at velocity v_i from the disk at position $z = 0$, at time $t = 0$ where the laser is started in time through the AOM for a duration t_{AOM} , to the entrance of the conical solenoid of the Zeeman slower at $d_z = 0.38$ m, (ii) a deceleration in the defined $B(z)$ profile (see figure 6) from v_i to $v[z(t_{AOM})] = v_i [1 - \alpha(z - d_z)]^{1/2}$ where $\alpha = [1 - (v_f/v_i)^2]/z_R$, $v_f = 252$ m/s and $z_R = 0.66$ m are calculus parameters, (iii) a free flight from $z(t_{AOM}) = v_i t_{AOM} - \alpha (v_i t_{AOM} - d_z)^2/4$ at constant slowed velocity $v(t_{AOM}) = v_i [1 + \alpha(d_z - v_i t_{AOM})/2]$ until eventually the crossing of ground state atoms of velocity v_0 that started at $T = 1$ ms from the second slot of the disk. Velocity v_0 is a priori unknown and is chosen somewhat arbitrarily being equal to 559 m/s and verified a posteriori through the time of flight data. The collision at relative velocity $v_r = v_0 - v(t_{AOM}) = v_i [\beta - 1 + \alpha(v_i t_{AOM} - d_z)/2]$ where $\beta = v_0/v_i$, experienced by the metastable atom in collision at the crossing point $z_c = (\beta v_i/v_r - 1)\beta v_i T + \alpha \beta v_i / (4v_r) [v_i^2 t_{AOM}^2 - d_z^2]$ with the ground state, allows two possible path from z_c until the detector at $z_{ch} = 2.385$ m. First choice is a “direct process” at $v(t_{AOM})$ after collision and a slowed atom detected at $t_{AOM} + (z_{ch} - z_c)/v(t_{AOM})$. A

second possibility is the loss of metastability at the crossing z_c and an “exchange process” giving a fast metastable atom at velocity v_0 detected at an earlier time $T + z_{ch}/v_0$.

Figure 5. Scheme of experimental set-up: a supersonic beam of argon atoms is excited to metastable states 3P_2 and 3P_0 by electron bombardment. At $z = 0$, the beam is sampled by a 3 slots (0° , 18° and 90°) rotating disk at 50 Hz. The laser is synchronously sent toward the beam in a 2 conical shape magnetic coils thanks to the 1st order transmitted AOM crystal, with the first slot at 0° for a duration t_{AOM} . At $z_c(t_{AOM})$ the 3P_2 slowed beam packet collides with the ground state beam packet coming from the 18° slot of the disk and continues at slowed down velocity its path until detection or continues at higher velocity v_0 in order to be detected earlier by the detector. Time of flight spectra are recorded at z_{ch} with the channeltron and at z_{DLD} with the delay line detector.

Figure 6. Magnetic field profile of the main Zeeman Slower:

Blue line: Zeeman-slower magnetic field in Gauss, versus distance z to the slotted disk. The entrance of the magnet is located at 0.25 m from the disk. The in-vacuum conical magnet is 0.80 m long. This curve is extrapolated from a measurement of the field with a Hall probe at 325 mA direct current (the coil has been used with 290 mA). The higher curve in magenta is a

model where formula $B(z)$ is deduced from $v(z)$ (see formula in the text) in order to merge the measured values of B in the range of interest $0.38\text{ m} < z < 1.18\text{ m}$.

II-2 Time of Flight (TOF) results

In one day of experiment, 24 TOF curves were recorded with the channeltron, using 14 mW of laser power in the following order: (i) a reference curve without laser, (ii) a curve with laser on permanently, (iii) a curve where the laser has permanently a residual power 0.7 mW facing the atomic beam (AOM off). Then AOM times are varied from 0.5 ms to 2.5 ms by steps of 0.1 ms. Each curve has 2000 points covering a range of 20 ms from the starting time (channel width 0.01 ms), then 20 000 sweeps are realized per spectrum. AS WELL 24 TOF curves are recorded with the DLD detector but with a lower temporal resolution of 0.05 ms.

First of all, we can make use of the UV photons created in the electron bombardment region through radiative decay. They are easily detected by the two detectors. Here, we get the channeltron spectra and do a counting statistic by adding the 2 firsts photon peaks at precise times 0 and 1 ms from the 24 acquisition curves, the third photon peak being drowned by the detected atomic signal is disregarded. We do get triangular shaped peaks of about 10 channels (0.1 ms) consistent with the convolution of two rectangular functions of about 0.05 ms equal width. This fact is verified by the 0.05 ms opening time of the disk's slots (1.55 mm large at a disk radius of 100 mm for a total disk diameter of 210 mm, frequency is 50 Hz). Assuming a detection radius through the inclined mirror of about 10 mm at 2.385 m from the disk position, a half divergence of 1.775 mrad can be inferred. This corresponds well with a UV point source located at the electron bombardment zone of about 437 mm from the disk position. Thus a total 3.5 mrad divergence can be assumed also for the atomic beam after the disk.

Looking at reference curve (see figure 7) allows determining an average measurement time of 4.30 ms from the photon peak maximum to the maximum of a well resolved metastable atom peak. This detection time corresponds well to the value 553 m/s at the peak of the velocity distribution function $g(v)$ defined here after and this with or without the convolution by the rectangular function. This fact has been checked since the measurement is the convolution of the centered rectangle function by a time modified Gaussian distribution defined from a velocity distribution of the form $g(v)=1/(\sqrt{2\pi}) \sigma \exp[-(v-v_i)^2/(2\sigma^2)]$. Then relative resolution in time and in longitudinal velocity coincides and is measured to be 8.3% FWHM. The magenta fitting curve matching the reference curve in figure 7 which is slightly asymmetric is found through the sum of two time-dependent modified gaussians: $f(t) = 5.47 \times 2/77 \times (z_{ch}/t)^2/(\sqrt{2\pi}) \times 18.5 \exp[-(z_{ch}/t-553)^2/(2 \times 18.5^2)] + 1.27 \times 2/77 \times (z_{ch}/t)^2/(\sqrt{2\pi}) \times 44 \exp[-(z_{ch}/t-558)^2/(2 \times 44^2)]$. It is then possible to extract the 3P_0 part of the metastable distribution through $1/6 f(t)$. Experimental temporal resolution is very good but counting is subjected to short and medium term fluctuations. Correction for count drifts will be done by normalizing to the area of the third peak of spectra.

3P_0 atoms are not affected by the laser and are evidenced though the first two peaks in the laser on curve of figure 7 in red. The tail just after the second peak and the beginning of the first peak testify of a small part of the 3P_2 atom distribution un-slowed and partially-slowed, but comparable in number to the 3P_0 distributions. Because the normally slowed distribution is clearly visible after the 3rd peak it is possible to extract from the laser curve corrected previously by a 1.25 factor because of counting drift, the signals presented in figure 8. Unfortunately the 3rd peak of the laser on spectrum being also affected by the slowing we still can normalize this spectrum with AOM curves that leave 3P_0 and residual 3P_2 signals naked.

It is possible to reconstruct the laser on signal by adding the 3P_0 contribution, the residual 3P_2 and the recognizable diffusive peak of fully slowed 3P_2 atoms. From figure 8, we can extract 105250 total counts from the laser on spectrum and from figure 7 we have 125878 total counts from the reference curve. It means that not all fully slowed 3P_2 atoms are detected and we can estimate their number to 20628. Thus we can give the following fractions of atoms: 3P_0 , 1/6 or 16.66%, 3P_2 is 5/6 or 83.33%. Among the 3P_2 population, 30% is un-slowed and badly slowed, 50% is a slowed detected population and then 20% is a slowed but undetected population. In conclusion 58% from a metastable peak are useful as 3P_2 atoms for slowing when the laser is on.

When the residual laser power is on the atomic beam, we do have a shift of the peaks showing than 3P_2 atomic distributions are affected. It is to remark that fast atoms are not hooked by the slowing process in the blue and red curve of figure 7. Their velocity can be estimated to be higher than 595m/s. Also the amount of atoms detected is smaller, sign of a general drift. This can be corrected at will by normalizing the spectrum to the 3rd peak of the reference signal.

Figure 7: black and magenta lines: reference time of flight signal without laser and a magenta fitted reference $f(t)$ (see text). Red line is a TOF un-corrected spectrum with laser on during the full acquisition. Notice the long tail after each of the 3P_0 peak, attributed to residual 3P_2 atoms not fully slowed. The blue line is a TOF un-corrected spectrum with the 0.7 mW residual power laser left through the AOM imperfectly “off”. Notice also the UV photon peak at 1 ms.

Figure 8: Time of flight spectrum with laser permanently on. The slowed 3P_2 distributions can be recognized by the steep walls at times 7.5 ms, 8.5 ms and 12.5 ms. 3P_0 graph in magenta is $1/6 f(t)$ (see text). Residual 3P_2 graph in blue is attributed to un-slowed and imperfectly slowed 3P_2 atoms.

II-3 Total indiscernible cross section results

In order to evidence the collision events, we propose to compare at some definite arrival times, slowed 3P_2 atoms from the first slot of the disk with slowed 3P_2 atoms from the second slot. We can do this because total numbers of counts from the first and second slit are very close to each other (see figure 7) and also because the time resolution 0.01 ms is very good with TOF results from the channeltron. Thus, we can do the ratio of slowed 3P_2 atoms at a predictable arrival time t_a in the steep wall of slowing spectra when t_{AOM} is ranging from 1.1 ms to 1.5 ms by steps of 0.1 ms. Clearly these atoms overlap the first half of the metastable atoms peak arriving from the second slot (supposedly with fast ground state atoms) and only affected by the residual power of the laser (see the 5 first arrows at the left of the graph in figure 9 at times in ms 4.80, 4.95, 5.10, 5.26 and 5.41). We can write this counting rate that we assume containing collisional information as $r_c = dN_c(t_a)/dt = L S n_0 n^* v_r \sigma$ where L is the collisional length of the two merging packets, S is the detected surface i.e. the surface of secondary electron emission formed by the inclined mirror, n_0 is the volume density of ground state, n^* is the slowed volume density of 3P_2 atoms and σ is the total indiscernible cross section defined in the previous theoretical part. Now, we look for atoms starting at time 1 ms from the second slot and slowed by the duration $t_{AOM} - 1$ ms (i.e. AOM spectra from 2.1 ms to 2.5 ms by steps of 0.1 ms). Their arrival times are the previous ones where we have to add exactly 1 ms and are represented by the next five arrows with the same colour code in figure

9. AS WELL, these times are very close and can be compared to the calculated ones following our slowing model in ms: 4.815, 4.95, 5.10, 5.254 and 5.417. This rate without collision is simply $r = dN(1+t_a)/dt = S n^* v^*$, where v^* is the slowed metastable velocity predicted by our model. Computing the measurable ratio of this two rates we get $R = L n_0 v_r/v^* \sigma$. We find respectively the values: 1.08, 1.4, 1.35, 1.39 and 1.08. AS WELL, we compute respectively the very comparable velocity ratios from $t_{AOM} = 1.1$ to 1.5 in ms to be equal to $v_r/v^* = 146/482 \sim 140/464 \sim 135/446 \sim 132/429 \sim 129/411 \sim 0.30$. We shall disregard time $t_{AOM} = 1.1$ ms (no collision is expected) and $t_{AOM} = 1.5$ ms (collision is far from the detector) since R is too close to 1. Selected 1.2, 1.3 and 1.4 ms AOM peaks will give respectively v_0 velocities: 600, 580 and 560 m/s and collisional energies 2, 1.9 and 1.8 meV for an average ratio $R = 1.38$. At 1.95 meV, the total cross section is computed to be $\sigma = 4954 a_0^2 \sim 1.39 \cdot 10^{-17} \text{ m}^2$. L is estimated by the gate duration $\delta t_d = 0.05$ ms of the disk. It equals at $t_a = 5.10$ ms: $L = z_{ch}/2 \times \delta t_d/t_a \sim 1.17$ cm. It is important to realise that this length is bigger than the longitudinal dimension of the slowed metastable collisional partner packet which is in the millimetre range, because the slowing effect is accompanied by cooling and then longitudinal compression in space, i.e. v^* is defined to $\delta v^* \sim v_{rec} \sqrt{N} \sim 1 \text{ m/s}$ with v_{rec} being the recoil velocity 1.2 cm/s and $N \sim 10000$ the number of absorbed photons. From these results we can infer the density of ground state atoms to be $n_0 \approx 2.86 \cdot 10^{13} / \text{cm}^3$.

If we assume an average velocity of ground state atoms $v_0 \sim 560 \text{ m/s}$ we find a flux density of $1.6 \cdot 10^{18} \text{ cm}^{-2} \text{ s}^{-1}$.

Figure 9 Normalized to third peak TOF spectra for increasing AOM durations. Thick red, blue and magenta arrows, are arrival times of colliding slowed atoms from the first slit of the disk with ground state atoms from the second slit of the disk for respectively $t_{AOM} = 1.2$ ms, 1.3 ms and 1.4 ms. Thinner red, blue and magenta arrows are slowed atoms for the same

AOM duration but from the second slit of the disk (no collision) and are obtained from spectra at $t_{\text{AOM}} = 2.2$ ms, 2.3 ms and 2.4 ms.

The same approach can be used with the DLD detector. The crossing being further at 2.995 m the collision energy will be smaller than with the channeltron results. We do find a ratio $R=1.3$ for the two AOM records 1.2 ms and 1.3 ms in figure 10. The collision energy is this time about 1.15 meV. As previously, we get now $v_r/v^* = 106.3/464 \sim 103/446.6 \sim 0.23$, $L \sim 1.19$ cm, $\sigma = 5817 a_0^2 = 1.6 \cdot 10^{-17} \text{ m}^2$ and, not too surprisingly, a close value for the ground state density $n_0 = 2.91 \cdot 10^{13} \text{ cm}^{-3}$.

Figure 10 Normalized to third peak TOF spectra for increasing AOM duration at the DLD detector. Thick red and blue arrows are arrival times of colliding slowed atoms from the first slit of the disk with ground state atoms from the second slit of the disk for respectively $t_{\text{AOM}} = 1.2$ ms and 1.3 ms. Thinner red and blue arrows are slowed atoms for the same AOM duration from the second slit of the disk obtained from spectra at $t_{\text{AOM}} = 2.2$ ms and 2.3 ms.

II-4 Partial indiscernible cross section results

It is also possible to extract the counts of metastable peaks from spectra with the channeltron or the DLD that are well separated from the unsloved peak coming from the second slot of the disk, with arrival times greater than 5.30 ms (see figure 11). These metastable atoms at slowed down velocity, have collided at z_c far from the detectors and the collection solid angle of detectors has to be taken into account. In this way, this contribution is related to partial indiscernible cross sections through

$$\sigma_{det} = \int_0^{\theta_{slow}} 2 \pi \sin\theta \left| \frac{f_g(\theta) + f_u(\theta) + f_g(\pi - \theta) - f_u(\pi - \theta)}{2} \right|^2 d\theta,$$

where $\theta_{slow} \approx \arctg \left[\left(1 - \frac{v_r}{v_i} \right) \frac{2 v_i}{v_r} \frac{\frac{\phi}{2\sqrt{2}}}{z_{ch} - z_c} \right]$ is the small center of mass acceptance angle of the detector (refer to paragraph I-2). We can write the total amount of slowed atoms from the first slit that collide at z_c and continue as slowed atoms as $N_{dir}^* = S L n_0 n^* v_r \sigma_{det}$. They can be compared to the full proportion of 3P_2 atoms subject to slowing $N^* = S n^* v^*$. The ratio $R^* = L n_0 v_r / v^* \sigma_{det}$ is deduced from measurement by the total amount of atoms in the “direct” peak divided by 63.3% of the total amount of atoms in the third peak (i.e. the useful 3P_2 population). It gives $R^* = 9069/14473$ for AOM duration 1.9 ms and 5930/12428 for 2.4 ms. It is to remark that the higher the AOM duration, the further the collision takes place from the detector and then metastable peaks show more losses at higher collision energies. Computing $L = z_c/2 \times \delta t_d / \langle t_a \rangle$ gives us 0.64 cm for $z_c = 1.583$ m and $t_a = 6.18$ ms for AOM duration 1.9 ms. AS WELL $L = 0.44$ cm for $z_c = 1.31$ m and $t_a = 7.5$ ms for the 2.4 ms AOM duration. Velocities v^* and v_r are respectively 341 and 218 m/s for 1,9 ms AOM and 253, 306 m/s for 2.4 ms AOM. If we use the density $n_0 = 2.9 \cdot 10^{13} \text{ cm}^{-3}$ we deduce the following partial cross sections: $\sigma_{det}(5\text{meV}) = 1888 \text{ a}_0^2$ and $\sigma_{det}(10 \text{ meV}) = 1105 \text{ a}_0^2$. We can compare these values to computed theoretical values 2100 a_0^2 and 1048 a_0^2 respectively for 5 and 10 meV. Total indiscernible cross sections are 4218 a_0^2 and 3540 a_0^2 . These results are summarized in figure 12.

Figure 11: TOF Spectra at AOM duration times 1.9 ms and 2.4 ms corresponding respectively to about 5 and 10 meV collision energies. 1.9 ms AOM result has 2 slowed peaks

that have collided: a main one at about 6.25 ms and a small residual peak at about 7.5 ms arrival times. 2.0 ms AOM spectrum shows a slowed peak at 7.5 ms. Higher peaks correspond to 0.9 and 1.4 ms duration time of slowing for $^3\text{P}_2$ atoms coming from the second slot of the disk (starting time 1 ms).

Following the same scheme « exchange » atoms should be in a $[\pi - \theta_{fast}, \pi]$ center of

mass domain where $\theta_{fast} \approx \arctg \left[\frac{2v_i}{v_r} \frac{\frac{\phi}{2\sqrt{2}}}{z_{ch} - z_c} \right]$. Partial cross section in this domain gives a 19 a_0^2 cross section at about 5 meV (AOM 1.9 ms). In term of counts this is less than 100 and absolutely not visible from the channeltron spectra. One could somewhat try to get them at the DLD position because they collide just in front of the mirror-channeltron first detector (at 2 meV, AOM = 1.4 ms) and they should be separated in time by a 0.35 ms visible delay at the DLD detector. This is also unsuccessful because their amount is not high enough to be evidenced from atoms coming from the second slot. It means also that at these energies “direct” cross sections dominate in all the spectra we did, at the channeltron AS WELL as the DLD detectors.

Figure 12 Total and partial indiscernible cross sections. Total cross section (full disk) are measured at 1 meV (at the DLD position) and at 2 meV (at the mirror position). They coincide reasonably well with theory (small square and guiding line). Partially solved cross sections are measured (full square) and calculated (small square and lower guiding line) showing similar results.

Finally, it should be noted that the knowledge of the ground state volume density would allow the measurement of the absolute value of the indiscernible cross section as a function of energy when varying the frequency of the TOF disk, that we did not do. For example reducing the frequency would allow to reach higher collision energies at the channeltron position but probably, it is going at high frequency of about 200 Hz that there is interest since cross sections get higher at lower energies ($E \approx 0.2$ meV is attainable) and might allow some “exchange” characterization at the DLD. Improvements will be feasible at not too much expenses: (i) working with no residual laser power at AOM off position (using a double passage through the AOM), (ii) having less residual 3P_2 atoms by increasing the available laser power and adjusting higher magnetic field for slowing, (iii) increasing the acquisition times to improve the statistics even more.

Conclusion

Using a double-slit chopping disk (a third slit being used as a reference) combined with a synchronous transient Zeeman slower, collinear collisions of metastable argon atoms with ground state argon atoms have been induced within the energy range 1 meV- 10 meV, explored for the first time to our knowledge. Since the collision partners are identical, the general problem of collision of indistinguishable particles arises in addition to that of the gerade-ungerade symmetry. It has been shown theoretically, using a new set of $\Omega = 2$ g and u molecular potentials, that this effect of identity of the partners, which makes intricate direct and exchange processes, actually strongly depends on the collision energy, becoming clear only either at large or at very low energies. It turns out that unfortunately (or fortunately?), the present energy range, combined with the collinear geometry in the lab reference frame, is such that the collision is strongly affected by *indistinguishability*, especially at large CM scattering angles. Nevertheless a satisfactory agreement between theory and experiment has been found. On another hand, almost pure exchange cross sections become measurable at lower energies, as shown *e.g* in [8,14] whereas at extremely low energy (ultra-cold regime) the collision $Ar^*(^3P_0 \text{ and } ^3P_2) + Ar$, governed by the s wave scattering, becomes purely elastic. The very large related total cross section [10] is a good test of the g potential, especially as concerns its asymptotic behaviour in C_6/R^6 .

Obviously the most direct way to eliminate all ambiguities coming from indistinguishability would be to use different isotopes. In the case of argon, the experiment is made difficult by the predominance of isotope Ar^{40} , while in principle feasible in so far as the deceleration could specifically concern, *via* the laser frequency, a rare stable isotope as Ar^{36} . On another hand, krypton atoms should allow us a much easier experiment for two reasons. First, they are well suited to our experimental setup (the laser wavelength is around 811.290 nm instead of 811.531 nm for argon). Second, several isotopes are present with significant populations in the natural mixture, allowing us to analyse AS WELL collisions of identical or different partners, in particular with isotope 83 whose abundance is 11.5 %, having a nuclear spin 9/2.

Acknowledgements

Authors are members of the French “Institut Francilien de Recherche sur les Atomes Froids”.

References

- [1] S. M. Trujillo, R. H. Neynaber and E W Rothe, Rev. Sci. Instrum. **37**, issue 12 , 1655 (1966)
- [2] V. A. Belyaev, B. G. Brezhnev and E. M. Erastov, Sov. Phys. JETP **52**, 1170-1177 (1967)
- [3] R. A. Phaneuf, C. C. Havener, G. H. Dunn and A. Müller, Rep. Prog. Phys. **62**, 1143-1180 (1999)
- [4] W. Vassen, C. Cohen-Tannoudji and M. Leduc, D. Boiron and C.I. Westbrook, A. Truscott and K. Baldwin, G. Birkel, P. Carnio, M. Trippenbach, Rev. Mod. Phys. **84**, 175-210 (2012)
- [5] B. M. Smirnov, Sov. Phys. JETP **19**, number 3, 692-696 (1964)
- [6] G. Hunter and Mary Kuriyan, Proc. R. Soc. Lond. A **358**, 321-333 (1977)
- [7] Jon P. Davis and W. R. Thorson, Can. J. Phys. **56**, 996, (1978)
- [8] B. D. Esry, H.R. Sadeghpour, E Wells and I Ben-Itzhak, J. Phys. B: At. Mol. Opt. Phys. **33**, 5329-5341, (2000)
- [9] B. Brutschy and H. Haberland, J. Phys. E **10**, 90 (1977)
- [10] D. Vrinceanu and H.R. Sadeghpour, Phys. Rev. A **65**, 062712 (2002)
- [11] P.D. Edmunds and P.F. Barker, Phys. Rev. Lett. **113**, 183001 (2014)
- [12] M.C. Castex and M. Morlais, F. Spiegelmann and J.P. Malrieu, J. Chem. Phys. **75** (10), 5006-5018 (1981) and F. Spiegelmann's Thesis 1983. Thèse d'Etat de l'Université Paul Sabatier de Toulouse (France)
- [13] C. Teichteil and F. Spiegelmann, Chem. Phys. **81**, 283-296 (1983)
- [14] E.R.T. Kerstel, C.P.J.W. Van Kruysdijk, J.C. Vlugter and H.C.W. Beijerinck, Chem. Phys. **121**, 211-235 (1988)
- [15] J-C. Karam, N. Wipf, J. Grucker, F. Perales, M. Boustimi, G. Vassilev, V. Bocvarski, C. Mainos, J. Baudon and J. Robert, J. Phys. B: At. Mol. Opt. Phys. **38**, 2691-2700 (2005)
- [16] J. Grucker, J. Baudon, F. Perales, G. Dutier, G. Vassilev, V. Bocvarski and M. Ducloy, J. Phys. B: At. Mol. Opt. Phys. **41**, 021001 (2008)
- [17] G.F. Gribakin and V.V. Flambaum, Phys. Rev. A **48**, 546 (1993)
- [18] J.-Y. Zhang, Z.-C. Yan, D. Vrinceanu, J. F. Bab, and H.R. Sadeghpour, Phys. Rev. A **73**, 022710 (2006)
- [19] Thierry Taillandier-Loize's PhD Thesis, 2014. Thèse de doctorat en physique de l'Université Paris 13 (France)
- [20] Handbook of mathematical functions, edited by Milton Abramovitch and Irene A. Stegun
- [21] B. Stern and J. Baudon, Z. Phys. D- Atoms, Molecules and Clusters, **8**, 359-365 (1988)
- [22] J. Robert, V. Bocvarski, I. Colomb de Daunant, G. Vassilev and J. Baudon, Journal de Physique **45**, 225-237 (1984)
- [23] N. F. Mott and H.S.W. Massey, The theory of atomic collisions, 3rd edition, Oxford at the Clarendon Press (1965 reprinted 1971)
- [24] S. Kasai, R. Mizutani, R. Kondo, M. Hasuo and T. Fujimoto, J. of the Phys. Soc. of Japan, **72**, n°8, pp. 1936-1942 (2003)
- [25] R.E. Grisenti, W. Schöllkopf, J.P. Toennies, G.C. Hegerfeldt and T. Köhler, Phys. Rev. Lett. **83**, 1755 (1999)
- [26] W.D. Phillips and H. Metcalf, Phys. Rev. Lett. **48**, 596 (1982)
- [27] E. L. Raab, M. Prentiss, A. Cable, S. Chu and D.E. Pritchard, Phys. Rev. Lett. **59** (23), 2631-2634 (1987)