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Differential measurements of metastable $\text{Ar}(3p^5 4s, {}^3P_{0,2})$ on $\text{Ar}(3p^6, {}^1S_0)$ collisions at thermal energies (*)

J. Robert, G. Vassilev, I. Colomb de Daunant, J. Baudon and V. Bocvarski (**)

Laboratoire de Physique des Lasers (***), Université Paris-Nord, avenue J.-B. Clément,
93430 Villetaneuse, France.

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Résumé. — On mesure les sections efficaces différentielles pour la collision $\text{Ar}^*(3p^5 4s, {}^3P_{0,2})$ sur $\text{Ar}(3p^6, {}^1S_0)$ aux énergies thermiques dans une expérience de jets croisés utilisant une méthode de temps de vol. On donne la dépendance angulaire de l'intensité diffusée, ainsi que la dépendance en vitesse de la section efficace pour le choc de front.

Abstract. — Differential cross-sections for $\text{Ar}^*(3p^5 4s, {}^3P_{0,2})$ on $\text{Ar}(3p^6, {}^1S_0)$ collisions at thermal energies have been measured in a crossed beam experiment, by a time-of-flight technique. The angular dependence of the scattered intensity and the velocity dependence of the differential cross-section in the backward scattering direction are given.

Differential cross-sections in metastable Ne on Ne collisions at thermal energies have recently been measured in supersonic [1, 2] or effusive [3, 4] crossed beam experiments. These measurements are a stringent test of the validity of the calculated Ne_2^* potentials, at intermediate or large inter-nuclear distances [5]. From a theoretical point of view, a comparison of Ne_2 and Ar_2 systems has now become possible [8], as *ab initio* calculations have recently been performed on the argon dimer [6]. It was therefore interesting to undertake differential measurements of metastable $\text{Ar}(3p^5 4s, {}^3P_{0,2})$ on $\text{Ar}(2p^6, {}^1S_0)$ collisions.

The experimental set-up shown in figure 1 is an adaptation of that described previously [3], where the effusive ground-state atom beam is replaced by a Campargue type supersonic beam. The main advantage of this beam is to be quasi-monokinetic ($\Delta v/v \lesssim 3\%$). Therefore it is no more necessary to chop it in order to select the ground-state atom velocity. The metastable atoms are produced by a pulsed electron bombardment of an effusive argon beam. The time-of-flight (T.O.F.) method allows then the study of a rather large range of relative energies ($35 \text{ meV} < E < 250 \text{ meV}$). In order to increase the time resolution, the scattered metastable particles are detected by a secondary electron emission from a metal plate perpendicular to the scattering direction, followed by a channel-electron multiplier. The detector has an acceptance solid-angle large enough (4.4×10^{-3} sterad.) to see the whole collision volume at all scattering angles.

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(**) Permanent address : Institut Za Fiziku, Studentski TRG 12 Beograd, Yugoslavia.

(***) Laboratoire associé au C.N.R.S. n° 282.

Fig. 1. — Experimental set-up. Metastable atoms are produced by a pulsed electron bombardment of an effusive Ar beam emerging from a capillary array. C is the collision volume. D is the secondary emission detector.

The number $N(\theta_L, t)$ of metastable atoms detected at an angle θ_L and after a time-of-flight t is proportional to :

$$\int_{-\infty}^{+\infty} dt_1 \frac{d\sigma}{d\omega_L}(v_r, \theta_L) \cdot v_r \cdot S(t_1) \cdot f\left(\frac{\tau_1}{l_1}\right) \cdot \left| \frac{\partial \tau_1}{\partial t_1} \right| \quad (1)$$

where $d\sigma/d\omega_L$ is the differential cross-section in the laboratory frame (averaged over the angular spread $\Delta\theta_L$), v_r is the relative velocity, $S(t_1)$ is the square gate function of the metastable atoms, $f(\tau_1/l_1)$ is the T.O.F. distribution [3] over the distance l_1 between the metastable atom source and the collision volume, and $\tau_1 = t - \frac{l'_1}{v'_1} - t_1 = \frac{l_1}{v_1}$ where v_1, v'_1 are respectively the velocities of the metastable atoms before and after the collision, l'_1 is the distance between the collision volume and the detector.

The differential cross-section $\sigma(\bar{v}_r, \theta_L)$ is then proportional to :

$$\frac{N(\theta_L, t)}{\bar{v}_r \int_{-\infty}^{+\infty} dt_1 S(t_1) f\left(\frac{\tau_1}{l_1}\right) \left| \frac{\partial \tau_1}{\partial t_1} \right|} \quad (2)$$

where $\bar{v}_r$ is the average relative velocity, calculated taking into account the finite time width ($\lesssim 20 \mu\text{s}$) of the metastable atom pulses $S(t_1)$; the corresponding energy resolution is then : $\delta E/E \lesssim 10\%$.

In order to avoid the effects of experimental drifts, over the relatively long time necessary to obtain T.O.F. spectra, we have measured their integral as a function of θ_L , keeping constant all parameters (electronic current, pressures, etc...). This gives the angular behaviour of the differential cross-section averaged over all present velocities (Fig. 2). It may be already seen on curve A, obtained with a limited angular resolution ($\Delta\theta_L \approx 3.2^\circ$), that the $\text{Ar}^*\text{-Ar}$ differential cross-section is very different from that of the $\text{Ne}^*\text{-Ne}$ collision at 78 meV [3] (curve C) : it is strongly peaked at

Fig. 2. — Average Ar*-Ar differential cross-section as a function of the lab. scattering angle θ_L . A : angular spread $\Delta\theta_L = 3.2^\circ$; B : $\Delta\theta_L = 1.2^\circ$; in fact the magnitudes given here by the curve A are smaller by a factor of 2 than the measured values. For sake of clarity, the curve A has been shifted to coincide at $\theta_L = 90^\circ$ with the curve B. C : Ne*-Ne differential cross-section. It has been also arbitrarily normalized at $\theta_L = 90^\circ$.

$\theta_L = 0^\circ$ and 90° . This character is even more marked when a better resolution ($\Delta\theta \approx 1.2^\circ$) is used (curve B). Moreover some undulations appear, particularly in the neighbourhood of 90° . It may be noticed that the secondary maximum seen at $\theta_L \approx 80^\circ$ in the Ne*-Ne differential cross-section corresponds to a rainbow effect due to $0_u(^3P_2)$ and $1_u(^3P_2)$ potential humps located at intermediate distance ($R \approx 5.5$ au). This feature is no more seen in the Ar*-Ar differential cross-section. This indicates that 0_u and 1_u potentials of this system present very low humps, or no hump at all, which is consistent with the *ab initio* calculations [6].

The resonant metastability-exchange process is predominant in the backward scattering region ($\theta_L \approx 90^\circ$). The T.O.F. spectrum obtained at $\theta_L = 90^\circ$ is shown in figure 3 (curve A). The first peak, at $t = 0$, comes from the U.V. photons emitted by the metastable atom source. It is followed by a narrow peak at $t = 200$ μ s, corresponding to metastable atoms created within the

Fig. 3. — Time-of-flight spectrum at $\theta_L = 90^\circ$. A : experiment ; B : normalizing factor $N_0(t)$ (see text) ; broken line : $\bar{v}_r/u$ (right hand scale) as a function of the time-of-flight, $\bar{v}_r$ is the relative velocity, u is the thermal velocity ($u = 353.5$ m/s).

supersonic beam by these resonant U.V. photons. Due to the finite time width of the metastable source pulses ($\approx 20 \mu\text{s}$) and to the angular aperture of the U.V. photon beam, this peak looks widened with respect to the actual T.O.F. width of the supersonic beam ($\approx 6 \mu\text{s}$). At larger times-of-flight, one observes the collisional T.O.F. spectrum, centred around $440 \mu\text{s}$. At this particular angle, the denominator in expression (2) becomes :

$$N_0(t) = \bar{v}_r \int_{-\infty}^{+\infty} S(t_1) f\left(\frac{\tau_1^D}{l_1}\right) dt_1 \quad (3)$$

where $\tau_1^D = t - t_1 - \frac{l_1'}{v_2}$. On the other hand, the time distribution of the incident metastable atoms at the collision point C is given by :

$$F_C(t) = \int_{-\infty}^{+\infty} S(t_1) f\left(\frac{\tau_1^C}{l_1}\right) dt_1 \quad (4)$$

where $\tau_1^C = t - t_1$. As v_2 is well defined (supersonic beam), one gets :

$$N_0(t) = \bar{v}_r F_C\left(t - \frac{l_1'}{v_2}\right). \quad (5)$$

The distribution $F_C(t)$ is directly measured by setting the detector plate at point C, and N_0 is then calculated (Fig. 3, curve B). The ratio $N(90^\circ, t)/N_0(t)$ leads finally to the $\bar{v}_r$ -dependence of the differential cross-section at 90° , averaged over the angular spread ($\Delta\theta_L \approx 3.2^\circ$) (see Fig. 4). As it is expected [7], this cross-section (dominated by the exchange contribution) is an increasing function of $\bar{v}_r$.

Fig. 4. — $\bar{v}_r$ -dependence of the differential cross-section at $\theta_L = 90^\circ$; the relative energies are given on the upper scale.

As it has already been shown in the case of the Ne_2^* system [4, 5], a more complete knowledge of the involved molecular states, as well as their mutual couplings, needs a detailed study of eventual inelastic processes (essentially the fine-structure transitions). This work is now in progress.

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