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THE ANGULAR DISTRIBUTION PARAMETERS OF ARGON, KRYPTON AND XENON FOR USE IN CALIBRATION OF ELECTRON SPECTROMETERS *

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Measurements are presented of the angular distribution parameter for the photoejected valence electrons of argon, krypton and xenon, from threshold to $h\nu \approx 25$ eV. The experimental arrangement at the NBS (SURF II) storage ring is described and the method of data analysis discussed. The results are compared with other experimental data and theoretical predictions.

1. Introduction

We present measurements of the asymmetry parameter β for photo-ejected valence electrons from the two spin-orbit-split levels in argon, krypton and xenon. The energy range studied extended from near the $^2P_{3/2,1/2}$ thresholds to approximately $h\nu = 25$ eV. Many measurements, both absolute [1-14] and relative [15,16], have been made of the angular distribution parameter of the outer p electrons of the rare gases, but the present data constitute the first detailed study close to threshold. Several of the earlier investigations suffered from experimental problems which led to a large variance in the β values, and it is only recently, with the advent of adequately prepared experimental arrangements, that reasonable agreement has been reached. Even so, these measurements have been restricted by the use of helium and neon line sources, and the data of Krause et al. [13] are the only other available measurements extending over a continuous photon energy range in which the two spin-orbit levels have been resolved. The agreement between the various sets of data generally becomes better as the photon energy increases; but near threshold, the region especially studied in the present experiment, serious discrepancies still persist.

Johnson and Cheng [17] have performed a multichannel photoionization calculation using the relativistic random-phase approximation for the outer shells of the rare gases, neon through to xenon. The present results may be used to test the theoretical predictions, particularly near threshold, where the interplay between correlation and relativity becomes more sensitive.

An important aim of the present esperiment was the production of reliable data for the calibration of future angular resolving electron spectrometers. Near threshold, for the rare gases studied, the asymmetry parameters are small, and as a consequence, become particularly sensitive to the

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state of polarization of the incoming photon beam and to stray fields. A calibration procedure is required to account for these effects and generate angular correction factors for the apparatus. The electron intensity at a specific angle is given by eq. (1) in section 2. The asymmetry parameter is calculated using the ratio of the electron intensities at any two angles. The angular correction factor, at a particular electron energy and collection angle, is the number by which it is necessary to multiply the intensity ratio in order to obtain the accepted β value. Therefore, the measurement of β in this region allows the determination of these experimental factors.

2. Experimental

The experiment was performed at the SURF II storage ring at the National Bureau of Standards, Washington. An angular resolving photoelectron spectrometer [18] was coupled to a high aperture 2 m normal incidence monochromator [19] incorporating a 2400 lines/mm osmium coated grating blazed at about 800 Å. The 0.1 mm high electron beam, constituting the entrance slit, resulted in a spectral resolution (fwhm) of 0.4 Å (corresponding to approximately 7 mV at 15 eV) and a photon flux of 5×10^{10} photons/s/bandwidth at 800 Å. The photoelectron spectrometer was a 50 mm mean radius hemispherical analyser and was operated in a constant pass energy mode of 5 V by the use of a pre-accelerating (pre-retarding) lens system. The analyser could be rotated in a plane at right angles to the incident monochromated photon beam with an angular acceptance of $\pm 2°$. The combined resolution of monochromator and electron analyser was approximately 120 mV in these experiments.

In the present experimental arrangement the light entering the interaction region was elliptically polarized and therefore the angular distribution of the electrons can be written in the convenient form [20]:

$$I(\theta) = k\left[1 + \tfrac{1}{4}\beta(3P\cos 2\theta + 1)\right], \qquad (1)$$

where $I(\theta)$ is the electron intensity at an angle θ to the major polarization axis (the plane of the electron orbit in the storage ring) and k incorporates unknowns such as the partial cross section, the atom number density and the electron analyser collection efficiency. The polarization of the light, $P = (I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp})$, was measured at each photon energy with a three mirror polarization analyser [18]. When $\beta = 0$, the electron intensity given by eq. (1) has no explicit dependence on P or θ. Therefore, the photon energy at which $\beta = 0$ allows the angular correction factor at that particular electron energy to be obtained directly.

At each photon energy electron spectra encompassing the two spin–orbit-split levels were collected at three angles, 0°, 45° and 90°, with respect to the major polarization axis. For krypton and xenon the two peak were completely resolved, and for argon they were easily deconvoluted by a computer program using the known spin–orbit splitting. The gas pressure in the interaction region was of the order of 10^{-3} Torr, giving an ambient pressure in the chamber of 5×10^{-5} Torr. Measurements were made as a function of gas pressure and no dependence of the asymmetry parameter was observed.

Calibration of the electron analyser with helium was not practical with the present experimental arrangement, so the measured relative β values had to be normalized. Recent values for the argon asymmetry parameter averaged for the $^2P_{3/2,1/2}$ levels, and measured at a few photon energies with the use of laboratory line sources, are within 5% agreement. By using these β values, together with the expectation of a smooth variation of β as a function of photon energy between the 3p ionization thresholds and the $3s3p^64p$ resonance at 26.62 eV, it was possible to obtain the angular correction factors as a function of electron energy. Many argon calibration runs have been performed over an extended time period to obtain the correction factors; the good agreement found for successive determinations leads to a high degree of confidence in their values. Also, transferring the calibrations so obtained to krypton and xenon resulted in good agreement with several accurate measurements and with the RRPA calculations, thus indicating on overall high degree of consistency and accuracy for all the rare gases.

An important aspect when reporting measurements that could be used for calibration purposes is a reasonable error estimation. The errors quoted in table 1 were derived from four sources: statistics, polarization, angular corrections and data reproducibility. The statistical error was generally small and only became significant at high photon energies, where the decreasing monochromator

Table 1
The present results of the asymmetry parameters for argon, krypton, and xenon

Wave length (Å)	Energy (eV)	Argon		Krypton		Xenon	
		$\beta_{3/2}$	$\beta_{1/2}$	$\beta_{3/2}$	$\beta_{1/2}$	$\beta_{3/2}$	$\beta_{1/2}$
950	13.05					0.54±0.12	
925	13.40					0.78±0.12	
900	13.78					0.95±0.11	
875	14.17					0.89±0.11	0.68±0.13
850	14.59			0.11±0.11		1.00±0.10	0.82±0.13
825	15.03			0.36±0.11		1.11±0.09	0.93±0.10
800	15.50			0.54±0.10	0.26±0.13	1.25±0.08	1.16±0.13
775	16.00			0.76±0.11	0.51±0.12	1.26±0.08	1.10±0.09
760	16.31	−0.17±0.11					
750	16.53	0.04±0.10	−0.03±0.13	0.75±0.08	0.55±0.10		
740	16.75	0.10±0.08	0.05±0.12				
736	16.85			0.81±0.10	0.80±0.14	1.37±0.08	1.16±0.09
730	16.98	0.12±0.08	0.11±0.11				
725	17.10	0.11±0.10	0.18±0.11	0.84±0.08	0.75±0.12		
720	17.22	0.21±0.08	0.17±0.10				
700	17.71	0.34±0.06	0.27±0.09	0.84±0.11	0.67±0.11	1.51±0.09	1.26±0.10
675	18.37	0.49±0.07	0.42±0.10	1.00±0.08	0.90±0.10	1.48±0.10	1.44±0.10
650	19.07	0.57±0.07	0.54±0.09	1.05±0.10	0.88±0.14	1.50±0.11	1.49±0.11
625	19.84	0.70±0.07	0.64±0.09	1.20±0.09	1.12±0.11		
600	20.66	0.78±0.08	0.69±0.10	1.24±0.09	1.10±0.12	1.65±0.12	1.59±0.10
584	21.23	0.86±0.07	0.81±0.11	1.26±0.11	1.13±0.13	1.55±0.14	1.65±0.12
575	21.56	0.96±0.09	0.95±0.11	1.27±0.11	1.22±0.14		
550	22.54	1.07±0.10	1.00±0.12	1.33±0.12	1.36±0.14	1.62±0.14	1.63±0.14
525	23.62	1.18±0.12	1.13±0.12	1.47±0.12	1.43±0.13		
500	24.80	1.16±0.13	1.19±0.13	1.67±0.13	1.61±0.14	1.77±0.15	1.59±0.15

output resulted in low electron count rates, and near threshold, where a combination of the electron spectrometer transmission efficiency and, to a lesser extent, the partial photoionization cross sections, again resulted in low count rates. Any error introduced by an incorrect value of the polarization P was particularly important for small but nonzero β values. In the main, however, the uncertainty in P was too small to affect β within the present degree of accuracy. The angular correction factor took into account any geometrical asymmetry in the interaction region; for example, any variation in the projected interaction region as seen by the electron analyser as a function of θ, and any magnetic or electric fields that could influence the electron trajectory. However, it should be stressed that the angular correction factor never amounted to more than approximately 4%. Finally, the β value for each spin–orbit-split level was determined using two of the three angular measurements and checked against the third. The difference between these two values, which was typically 3–4% and never exceeded 7%, acted as a guide to possible systematic errors.

3. Results

3.1. Argon

The present results for the β parameter of the two spin–orbit-split levels of the argon ion are plotted in fig. 1, together with other recent experimental data and the theoretical curves of Johnson and Cheng [17]. Previous caluclations, by Amusia et al. [21] and Taylor [22], only predict values for an unresolved β. Owing to the good agreement amongst the many measurements carried out at 584 Å, only a few selected points have been plotted. A detailed study of the asymmetry parameters for argon, krypton and xenon at 584 Å, and comparison of all previous data, has been described by

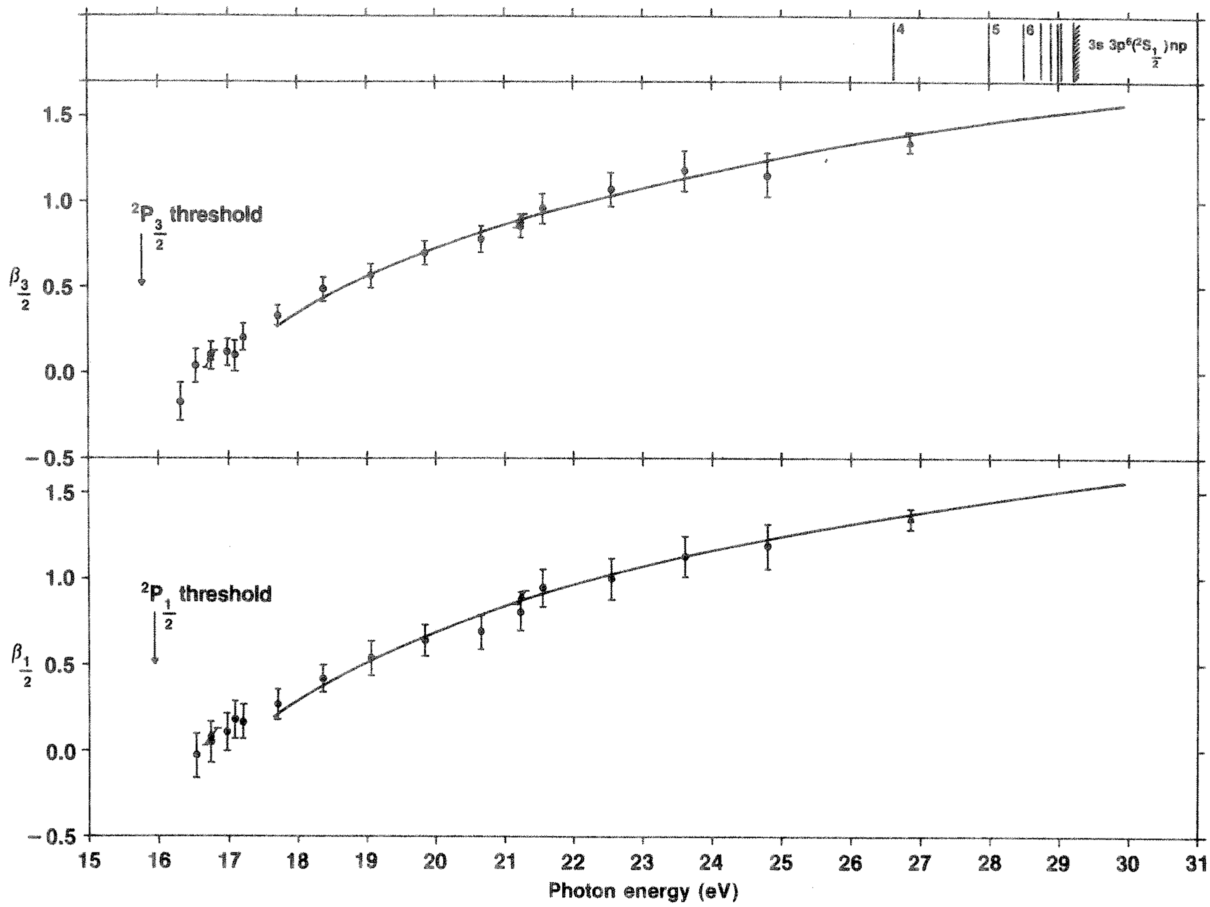

Fig. 1. The argon asymmetry parameters. Present results, ●; Dehmer et al. [9] at 16.67/16.85, 21.22 and 26.86 eV, ▲; Johnson and Cheng [17], solid line.

Kreile and Schweig [12]. Also shown in fig. 1 are the positions of the $3s3p^6(^2S_{1/2})np$ resonances [23]. The lowest energy two electron resonance lies at 29.04 eV. The photoionization cross section between the $^2P_{3/2,1/2}$ thresholds, at 15.76 and 15.94 eV respectively, and the $3s3p^6(^2S_{1/2})4p$ resonance at 26.62 eV is smooth, and therefore it is to be expected that the two β components will exhibit a similar smooth form. In an earlier investigation using the present apparatus, Codling et al. [24] studied the variation of β through the $3s3p^6(^2S_{1/2})4p^1P_1^0$ resonance, and the present data fit smoothly into the wings of the previous measurement. Some measurements which were taken near threshold indicated a sharp fall in the value of β for both the $^2P_{3/2,1/2}$ levels. However, since the emphasis of the present study was the collection of reliable data for calibration purposes, these points have not been plotted, due to the uncertainty in the angular correction factor for electron energies of less than 300 to 400 meV. The multichannel photoionization calculation, using the relativistic random-phase approximation by Johnson and Cheng [17], shows good overall agreement with the present results, although there is an indication that the calculation would predict lower values for both β components in the threshold region than have been observed. It would be of particular interest to extend these calculations to lower energy to find the theoretical value at which β becomes zero since, as already discussed, this can yield important experimental information about the angular correction factor.

3.2. Krypton

Fig. 2 shows the present β results for krypton, together with the experimental data of Dehmer et

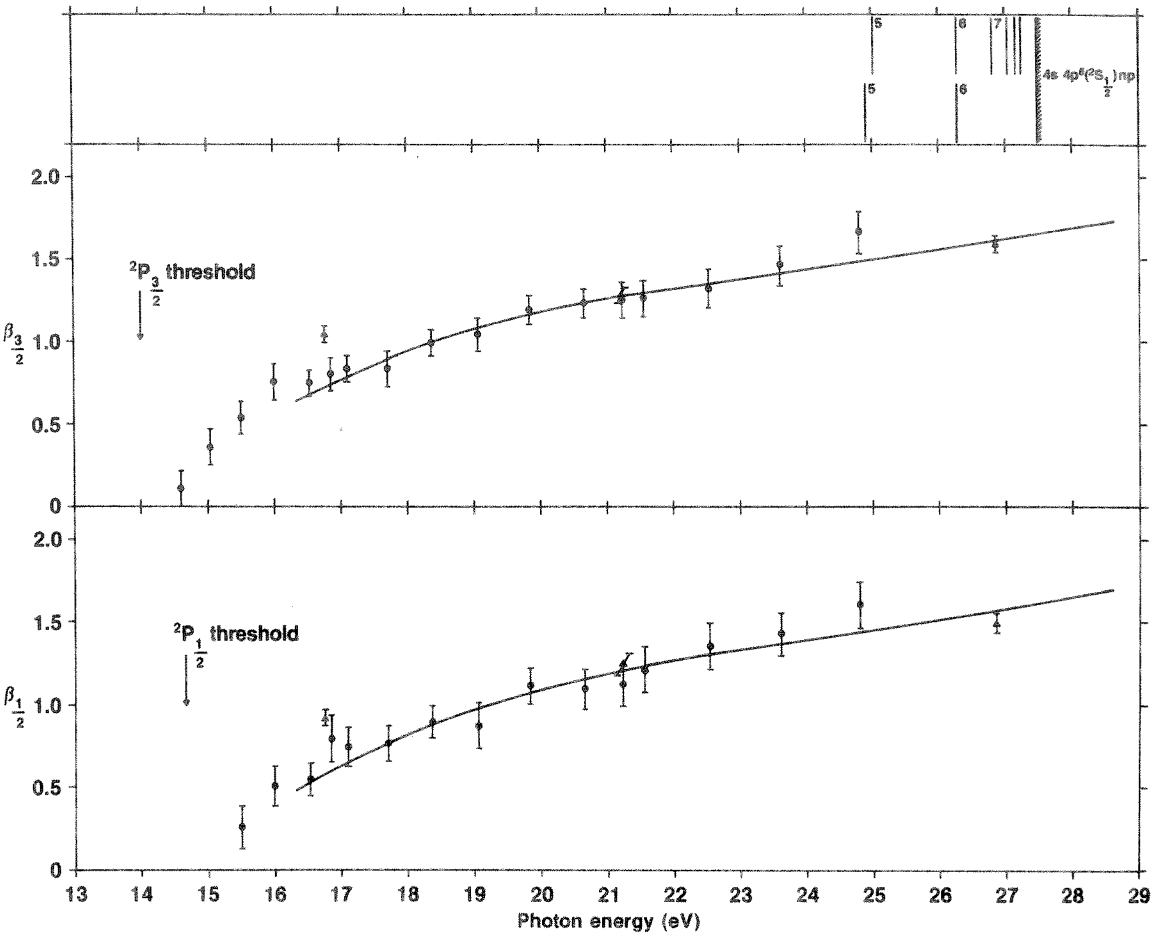

Fig. 2. The krypton asymmetry parameters. Present results, ●; Dehmer et al. [9] at 16.67/16.85, 21.22 and 26.86 eV, ▲; Johnson and Cheng [17], solid line.

al. [9] and theoretical results [17]. Very good agreement is attained at 584 Å between the present and most of the recent experimental results, so not all of the available data could be plotted. As with argon, the krypton asymmetry parameters at 584 Å have been reviewed thoroughly by Kreile and Schweig [12]. There appears to be a serious discrepancy between the present data and those of Dehmer et al. [9] taken with the neon 736/744 Å resonance lines. Their values lie significantly higher than the present results. Since the krypton $^2P_{3/2,1/2}$ ionization thresholds lie at 14.00 and 14.67 eV, respectively, the photoelectrons are ejected with sufficient energy for the correction factors used in the present experiment to be determined accurately. In addition, no structure occurs in the total photoionization cross section between the ^2P thresholds and the $4s4p^6(^2S_{1/2})5p$ resonances [25] that could account for any sudden variation in β. The lowest energy two electron transitions occur in the region of the lowest member of the $4s4p^6(^2S_{1/2})np$ Rydberg series and may possibly perturb the monotonic increase in β. The agreement with the theoretical result [17] is generally good, apart from the low energy, near-threshold region and at 24.80 eV. The discrepancy with the high energy data point may be explained by the nearness of the $4s4p^6(^2S_{1/2})5p$ resonances at 24.92 and 24.99 eV [25]. The variation of β through this resonance region will be the subject of a forthcoming publication [26]. Again, some β measurements which were taken at photon energies close to threshold have not been shown, due to the uncertainty in the correction factors, but would suggest a sharp decrease for both components.

3.3. Xenon

The asymmetry parameters for the two spin–orbit-split levels of the xenon ion are shown in fig. 3. The overall agreement between the present data and other experimental and theoretical values is good. Several of the Rydberg series [25] due to one or two electron excitation, and which occur within the energy range of interest, are indicated in the figure, but do not appear to have influenced the present data. The β value at 21.23 eV for the $P_{3/2}$ level lies somewhat lower than the other experimental results and the theoretical predication. The $5s5p^6(^2S_{1/2})6p\ ^1P_1^0$ resonance occurs at 20.95 eV and Codling et al. [24] have studied the variation of β through a part of this region. The β values obtained in their investigation merge with the present results and with an improved study in the resonance region by Ederer et al. [26]. As discussed previously for the case of krypton, the lack of structure in the total photoionization cross section between the $^2P_{3/2,1/2}$ thresholds at 12.13 and 13.44 eV respectively, and the low lying members of the indicated Rydberg series, would tend to suggest that a smooth variation of β as a function of energy would be expected. In addition, the RRPA calculations [17] predict a smooth form and the present data confirm such a trend. The agreement with the only other synchrotron radiation results by Krause et al. [13] appears satisfactory, although the region of overlap is not large. As in the cases of argon and krypton, some omitted results near threshold would suggest a fairly sharp fall in the β values for both components.

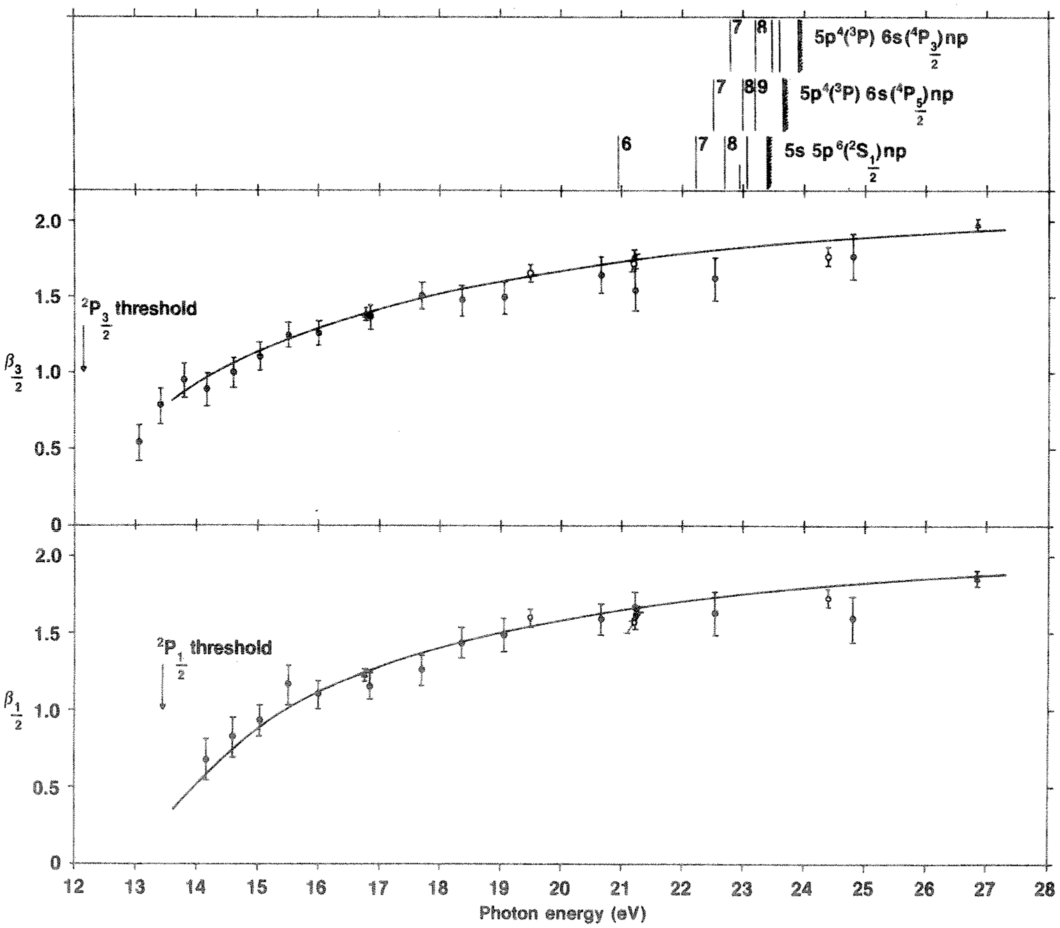

Fig. 3. The xenon asymmetry parameters. Present results, ●; Dehmer et al. [9] at 16.67/16.85, 21.22 and 26.86 eV, ▲; Krause et al. [13] at 19.5, 21.2 and 24.4 eV, ○; Johnson and Cheng [17], solid line.

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