POLARIZATION MEASUREMENTS IN He^+ - N_2 COLLISIONS

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ABSTRACT. Emission cross-section and linear polarization of the excitation of helium atomic HeI (388.9nm) and nitrogen ionic NII (500.1-500.5nm) lines have been measured in a broad range of collision energy (1-10keV) of He^+ ions. High degree of the polarization P = - 20% was observed in the case of helium line. Such a great negative value of the degree of polarization indicates that $m_L = \pm 1$ sublevels of the excited state 3^3P of helium atom are preferably populated. Analysis of the experimental results indicates that the electron density formed in He^* during the collision is oriented perpendicularly with respect to the incident beam direction. Strong correlation is revealed between inelastic channels of the formation of excited helium and nitrogen particles.

INTRODUCTION

Ion-impact processes on molecular nitrogen N_2 have been studied extensively because of its importance in many natural and applied phenomena. From the practical point of view the collision that produces electronically excited species plays a key role in plasma physics, gas discharges, in the study of the interstellar medium and in the upper layers of the atmosphere.

Numerous works have been devoted to investigate the polarization of radiation in ion-atomic and ion-molecule processes [1-13]. Such investigations aim to determine electron alignment and polarization effects [4-9]. The polarization fraction is quantitatively discussed in terms of alignment of the orbital momentum sublevels. The cross sections of population of magnetic sublevels provide detailed information on the excitation mechanism. Due to the different populations of magnetic sublevels within a certain (nl) subshell, the radiation can be polarized and, consequently, anisotropic. Hence, from experimental point of view the information about the polarization is important for the determination of accurate absolute and relative photon emission cross sections.

Usually in the polarization measurements the coincidences of photon and scattered particle are detected. In the above mentioned works the Lyman-α line of the hydrogen atom was mainly registered. In order to amplify the optical registration sensitivity instead of monochromator the broad bandpass filters have been used for isolating the optical lines.

In case of $He^{+} - N_2$ colliding system the radiation spectrum is quite multitudinous and therefore the monochromator with high resolving power (~ 0.2 nm) is to be used. As a result the optical sensitivity became worse and it was impossible to use the coincidence scheme in measurement. The obtained results of polarization experiments allowed to make some nontrivial conclusions about the spatial distribution of the electron cloud.

In this paper, we report experimental results of the study of the excitation processes in ion-molecular collisions in a broad range of collision energy (1-10 keV). In the visible range (380 ÷ 800 nm) we have measured excitation functions and degree of linear polarization for the lines of the helium atom (λ = 388.9nm, transition 3p $^3P^0 \rightarrow$2s ^3S)) and nitrogen ion (λ = 500.1÷500.5nm, transition 3d $^3F^0 \rightarrow$3p ^3D) due to the He^+-N_2 collision.

APPARATUS AND METHOD OF MEASUREMENT

The experimental setup and calibration procedure have been described in details in [13]. The ion beam extracted from the discharge source is focused, accelerated and mass selected in a 60^o magnetic sector field. The beam of He^+ was passed through collision chamber. The radiation emitted as a result of the excitation of colliding particles was observed at 90^o to the direction of the beam. The spectroscopic analysis of the emission was performed with visible monochromator incorporating the diffraction grating with resolution - 40 nm/mm.

Polarizer and the mica quarter-wave phase plate are placed in front of the entrance slit of the monochromator and the linear polarization of the emission is analysed. The phase plate was placed after the polarizer, was rigidly coupled to it, and applied to cancel the polarizing effect of the monochromator. The emission was recorded by photomultiplier with a cooled cathode and operated in the current mode.

The helium ion currents in the collision chamber were of the order 0.1-0.5μA and the pressure of the gas under investigation did not exceed $6 \cdot 10^{-4}$ Torr, so that multiple collisions could be ignored. The system was pumped differentially by the oil-diffusion pump. The residual-gas pressure did not exceed $0.1 \cdot 10^{-6}$ Torr.

Particular attention was devoted to the reliable determination and control of the relative and absolute spectral sensitivity of the light-recording system. This was done by measuring the photomultiplier output signal due to the (0.0), (0.1), (0.2), (0.3), (0.4), (1.2), (1.3) and (1.4) bands in the first negative system of the ion N_2^+ (B $^2\Sigma_u^+$- - X $^2\Sigma_g^+$ transition) and (4.0), (4.1), (6.2), (6.3), (2.0), (3.0), (5.1) and (5.2) bands of the Meinel system (A $^2\Pi_u$-X $^2\Sigma_g^+$ transition) excited in collisions between the (E_e=110 eV) electrons and nitrogen molecules. The electron gun was located directly in front of the entrance slit of the collision chamber. The output signal was normalized to (0.1) band (λ = 427.8 nm), which had the high intensity in this range. The relative spectral sensitivity curve of the recording system obtained in this way was compared with the relative excitation cross sections for the same bands, averaged over the experimental data reported in ref. [14-18]. The absolute excitation cross-sections for the (0.1) band (λ = 427.8 nm) were assumed to be $5.3 \cdot 10^{-18}$ cm^2 at the electron energy of 110 eV. This value was taken from ref. [14].

The relative uncertainty in our measurements was 5%, the absolute uncertainty was 15%. The accuracy of polarization measurements did not exceed ~ 2%.

RESULTS OF MEASUREMENTS AND DISCUSSION

Excitation functions measured for lines of the helium atom HeI (λ=388.9nm, 3p ^{3}P→2s ^{3}S) and nitrogen ion NII (λ = 500.1÷500.5nm, 3d ^{3}F→3p ^{3}D) are plotted in Fig.1. Presented curves exhibit surprising resemblance: in the whole investigated energy region both the absolute values of the emission cross sections and their energy-dependence are close to each other.

Results of polarization measurements are presented in Fig.2. As shown maximum degree of polarization is 20% for HeI (388.9 nm) line and that is ~ 5% for NII (500.1÷500.5 nm) which is a dissociation product. It is clear from Fig.2 that degrees of polarization for the investigated emission lines change the sign at the nearly same energy ~3 keV and reach their maximum in region of 8-10 keV of the incident He^+ energy. The obtained results presented in

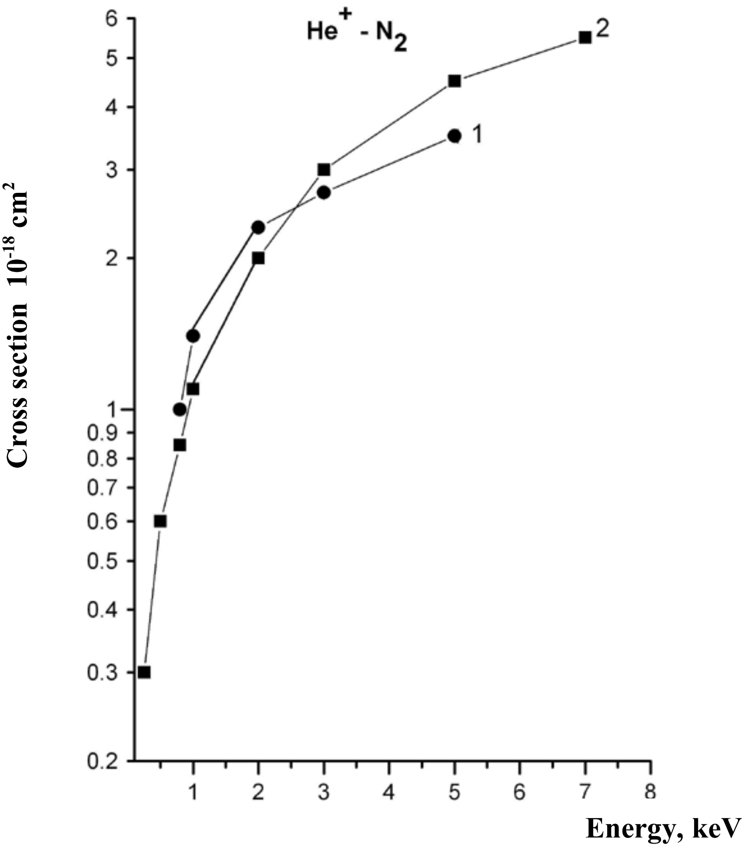

Fig.1. Energy dependence of the excitation cross section of helium atomic and nitrogen ionic lines: 1- HeI(388.9 nm), 2 -NII (500.5 nm)

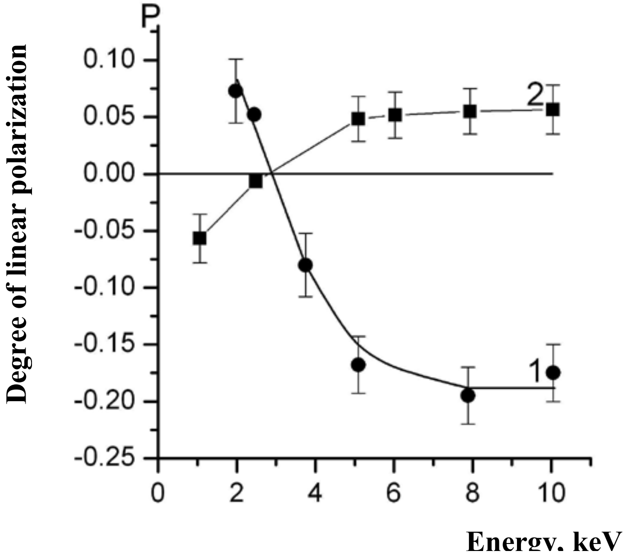

Fig.2. Energy dependence of the degree of polarization
1 - HeI(388.9nm), 2- NII(500.5nm)

Figs.1 and 2 point to the fact that there should exist a strong correlation between inelastic processes producing the excited helium atoms and nitrogen ions (dissociation products)

$$He^{+} + N_2\left(X^1\Sigma_g\right) \rightarrow He^{*} + N^{+} + N \qquad (1)$$

Energy, keV

$$He^{+} + N_2\left(X^1\Sigma_g\right) \rightarrow He + \left(N_2^{+}\right)^{*} \qquad (2)$$

Also we have supposed that the inelastic energy defects for these channels are close to each other.

Polarization of the emission emerging from excited ^3P-state of helium is connected to the relative populations of $m_L = 0$ and

$m_L = \pm 1$ sublevels. Expression for the first Stock's parameter has been derived on the basis of general approach developed by Macek and Jaecks [1]. We remove the details of these calculations into Appendix presenting here only the final formula for linear polarization:

$$P = \frac{I_{//} - I_{\perp}}{I_{//} + I_{\perp}} = \frac{15(\sigma_0 - \sigma_1)}{41\sigma_0 + 67\sigma_1}, \quad (3)$$

where $I_{//}$ and $I_{\wedge}$ denote measured intensities of radiation with electric vector parallel and perpendicular to the incident ion beam, respectively; σ_0 and σ_1 stand for the cross sections for population of sublevels with $m_L = 0$ and $m_L = \pm 1$, respectively. Taking into account that experimentally observed value of *P* is 20% one obtains from (1) that $\sigma_1/\sigma_0 \approx 15$. Such a great value of this ratio indicates that $m_L = \pm 1$ sublevels of the excited helium atom are preferably populated. This means that the electron density formed in He^* during the collision is oriented perpendicularly with respect to the incident beam direction.

For polarization of radiation emitted by nitrogen ion N^+ (dissociation product) we have used the same technique as for derivation of (3) and the following expression has been obtained:

$$P \approx \frac{\sigma_0 + 2\sigma_1 - 3\sigma_3}{3\sigma_0 + 6\sigma_1 + 6\sigma_2 + 5\sigma_3} \quad (4)$$

We note that in case of excited N^+-ion it is complicated to trace any pronounced alignment of the radiating object. The reason is that expression (4) contains not only σ_0 and σ_1, but σ_2 and σ_3 too and the unambiguous determination of branching ratios seems to be a difficult task.

There are some additional arguments that substantiate the existence of correlation between the channels (1) and (2). Filippelli et. al in ref. [19] investigated electron impact dissociative ionization of N_2. The authors have observed the same emission line of N^+ -

500.5nm. Because of the small mass of the incident particle – electron, the threshold impact energy for the appearance of this line nearly coincides with the corresponding energetic defect less than the helium atom ionization potential. So, 56eV for the threshold after subtraction of 24.6eV gives approximately 32eV for the energy defect. In the energy loss spectrum plotted by Dowek et al. [20], for the charge exchange channel, one can find a broad peak in the energy range of ~ 30 eV. We see that 32 eV is located in this area and this fact is indirect evidence in favor of the close relationship between reactions (1) and (2).

The energy dependence of measured polarizations shows that the electronic orientation of the excited He atom changes at nearly 3keV. One can suppose that because of the mentioned strong correlation between the channels of excitation of He and N^+ the electronic orientation of the excited nitrogen ion would also change. This implies that the effect of molecular axis orientation with regard to incident ion beam is also changed as energy increases.

APPENDIX

Here we derived the simple formula for the degree of polarization of radiation as result of the atomic particle collision process. Presented calculations are based on the pioneering work of Macek and Jaecks [1]. Below we use the following quantum numbers: L-electronic orbital quantum number, M_L-magnetic quantum number, J-full electronic momentum quantum number, *S*- electronic spin quantum number, I- nuclear spin quantum number and F – full atomic momentum (electronic + nuclear) quantum number.

In the polarization experiments number of the photon and projectile coincidences dN_c depends upon the position of the photon and particle detectors. The incoming beam axis is usually taken to be the z-axis, and the *x*-z plane is located arbitrarily. The angular coordinates of the particle detector relative to this coordinates system are denoted by θ and φ, and the coordinates of the photon detector by θ' and φ'. Number dN_c is proportional to linearly polarized light intensity which is oriented at an angle β with respect to z-axis, provided projectile particle is scattered in (θ, φ) direction.

$$dN_c = \{A_{00}\cos^2\beta + A_{11}\sin^2\beta + (A_{11} - A_{00})\cos^2\beta\cos^2\theta +$$
$$+\sqrt{2}\mathrm{Re}A_{01}\left[\sin2\theta'\cos^2\beta\cos(j - j') + \sin2\beta\sin\theta'\sin(j - j')\right] -$$
$$-\mathrm{Re}A_{1\ -1}\left[(\cos2\beta\cos2\theta' - \sin^2\beta)\cos2(j - j') +\right.$$
$$\left.+\sin2\beta\cos\theta'\ \sin2(j - j')\right]\}\, d\Omega d\Omega' \quad \text{(A1)}$$

$d\Omega$ and $d\Omega'$ are the solid angles covered by the particle and photon detector, respectively. Coefficients $A_{qq'}$ are determined as:

$$A_{qq'} = \sum_{JFJ'F'M'_L M_L} U(qq'M_L M'_L JFJ'F'LL_0)\left\langle a_{M'_L} a_{M_L}\right\rangle \int_0^{\Delta t} dt e^{-(\gamma + i\omega_{JFJ'F'})t} . \quad \text{(A2)}$$

Here

$$U(qq'M_L M'_L\, JFJ'F'LL_0) = \quad \text{(A3)}$$

$$= \frac{(2J+1)(2J'+1)(2F+1)(2F'+1)(2L+1)}{(2S+1)(2I+1)}(-1)^{L_0+q-M_L}\sum_{\chi=0,1,2}(2\chi+1)(-1)^{\chi}$$

$$\times\begin{Bmatrix} L & L & \chi \\ J' & J & S\end{Bmatrix}^2\begin{Bmatrix} J & J' & \chi \\ F' & F & I\end{Bmatrix}^2\begin{Bmatrix} L & L & \chi \\ 1 & 1 & L_0\end{Bmatrix}\begin{pmatrix} L & L & \chi \\ -M'_L & M_L & \nu\end{pmatrix}\begin{pmatrix} 1 & 1 & \chi \\ -q & q' & -\nu\end{pmatrix},$$

q is polarization vector component of the photon; ω is the frequency of the emitted light and $1/\gamma$ is the mean life of the excited atom. The excitation amplitudes contain all information about collision dynamics and they depend on θ only. Time integration in (A2) involves detection time interval 0-Δt.

In our experiment we do not fix scattered particles. This means that expression (A1) should be integrated over coordinates θ and φ. Furthermore, in our experimental condition the photon detector is installed in direction perpendicular to the primary ion beam, i.e. $\theta' = 90^0$. As to analyzer angle β, it was taken equal to 0^0 and 90^0. Therefore only the following terms will contribute to the detected intensity

$$I \sim (A_{00}\cos^2\beta + A_{11}\sin^2\beta)d\Omega' \qquad \text{(A4)}$$

In case when radiation from helium atom is observed, nuclear spin I = 0, so we have no hyperfine structure. Consequently, we can change $\omega_{JFJ'F'}$ by $\omega_{JJ'}$. Further, since the mean life of excited atom $1/\gamma$ and $1/\omega_{JJ'}$ is much shorter than commonly employed resolution time, the time integral becomes:

$$\int_0^{\infty} dt\exp[-(\gamma + i\omega_{JJ'})t] = \frac{1}{\gamma + i\omega_{JJ'}} \approx \begin{cases} 0\ , & J \neq J' \\ 1/\gamma\ , & J = J' \end{cases}. \qquad \text{(A5)}$$

Now, (A1)-(A5) allow to find an exact value for the first Stocks parameter

$$P = \frac{I(\beta = 0^\circ) - I(\beta = 90^\circ)}{I(\beta = 0^\circ) + I(\beta = 90^\circ)}. \qquad \text{(A6)}$$

Let's determine degree of polarization for He atom line (388.9 nm, transition ${}^3p \to {}^3s$). For this case when q = 0 q′= 0,

$$U(00M_L M'_L\ JJJJ10) = (2J+1)^4(-1)^{-M_L} \sum_{\chi=0,1,2}(2\chi+1)(-1)^{-\chi} \times$$

$$\times \begin{Bmatrix} 1 & 1 & \chi \\ J & J & 1 \end{Bmatrix}^2 \begin{Bmatrix} J & J & \chi \\ J & J & 0 \end{Bmatrix}^2 \begin{Bmatrix} 1 & 1 & \chi \\ 1 & 1 & 0 \end{Bmatrix} \begin{pmatrix} 1 & 1 & \chi \\ -M_L & M_L & 0 \end{pmatrix} \begin{pmatrix} 1 & 1 & \chi \\ 0 & 0 & 0 \end{pmatrix},$$

and when q = 1 q′ = 1,

$$U(11M_L M'_L\ JJJJ10) = (2J+1)^4(-1)^{1-M_L} \sum_{\chi=0,1,2}(2\chi+1)(-1)^{-\chi} \times$$

$$\times \begin{Bmatrix} 1 & 1 & \chi \\ J & J & 1 \end{Bmatrix}^2 \begin{Bmatrix} J & J & \chi \\ J & J & 0 \end{Bmatrix}^2 \begin{Bmatrix} 1 & 1 & \chi \\ 1 & 1 & 0 \end{Bmatrix} \begin{pmatrix} 1 & 1 & \chi \\ -M_L & M_L & 0 \end{pmatrix} \begin{pmatrix} 1 & 1 & \chi \\ -1 & 1 & 0 \end{pmatrix}$$

Here ( ) and { } denote the 3-j and 6-j symbols, respectively.

Finally, we obtain the following expression for the degree of polarization for the mentioned helium emission line:

$$P = \frac{15(\sigma_0 - \sigma_1)}{67\sigma_0 + 41\sigma_1}, \qquad (A7)$$

where σ_0 and σ_1 are cross-sections of population of magnetic sublevels with $m_L = 0$ and $m_L \pm 1$, respectively.

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