Electron spectra from slow collisions of excited noble gas atoms

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Electron spectra arising from two-body collisions of excited noble gas atoms are analyzed. The Penning- and associative-ionization channels for the He(2^3S)–He(2^3S), He(2^3S)–He(2^1S), and Ne($^3P_2$)–Ne($^3P_2$) reactions are separated by the plasma electron spectroscopy method for the first time.

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§1. INTRODUCTION

Ionization processes taking place in collisions of two excited noble gas atoms in radiative or metastable states,

$$A^{*}+B^{*}\rightarrow\begin{cases}A+B^{+}+e, & (1)\\ A^{+}+B+e, & \\ AB^{+}+e, & (2)\end{cases}$$

are of interest not only in themselves, but also in connection with applications, since these processes can substantially affect the properties of a plasma.[1–3] Until recently, the comparatively few studies of reactions (1) and (2) were based on experiments with a plasma afterglow or with merging beams and were limited[1,2] to the study of the energy dependence of the cross section, $\sigma(E)$, or the temperature dependence of the rate constant, $K(T)$. Mass-spectrometer measurements of the relative fraction Q of molecular ions have been made for four reactions, but, as reported in the review by Neynaber,[2] the data obtained for the He(2^3S)–He(2^3S) reaction obtained in different studies differ by more than an order of magnitude.

More complete information on processes (1) and (2) could be obtained using Penning electron spectroscopy (see, e.g., Ref. 3), but, because of the obvious difficulties in experimenting with colliding excited atoms, such studies have not yet been carried through. Moreover, one must know the basic features of the electron spectra in order to interpret them. A computer calculation of the spectrum for the He(2^3S)–He(2^3S) reaction at $E = 0.03$ eV has been published[4] but it is impossible to determine how characteristic the spectrum is without analyzing its features; moreover, what is most important, neither the overlap of the electron spectra arising from reactions (1) and (2), which is of vital importance for the practical application of Penning electron spectroscopy, nor the effect of the velocity distribution of the atoms, were discussed in Ref. 4. These problems will be discussed in § 2.

In § 3 we propose a new method for investigating electron spectra—plasma electron spectroscopy (PLES)—which is based on the study of the electron energy distribution function in a decaying plasma and on the properties of the electron spectra established in § 2. The method combines the possibility of achieving a high concentration of excited atoms in a plasma afterglow with the advantages of Penning electron spectroscopy. Using PLES we have determined Q for the He(2^3S)–He(2^3S) reaction and, for the first time, for the He(2^3S)–He(2^1S) reaction, and have obtained an upper bound for Q for the Ne($^3P_2$)–Ne($^3P_2$) reaction.

§2. DETERMINATION OF THE CONTRIBUTION FROM MOLECULAR IONS TO THE ELECTRON SPECTRUM

In studying ionization processes it is of interest to determine the ratio Q, either for the cross section or for the associative ionization (AI, reaction (2)) rate constant, to the total cross section or the rate constant for reactions (1) and (2). Q can be determined if reactions (1) and (2) result in the release of electrons having substantially different energies, in other words, if the overlap of the AI spectrum and the Penning ionization (PI) spectrum is small as compared with the total width of the entire spectrum. The characteristics of the ionic and excited-state terms of the noble gas atoms are such that the high-energy part of the spectrum will be mainly due to AI, and the low-energy part, to PI. Let us consider the overlap of the AI and PI spectra in more detail.

The basic principles for calculating reactions such as (1) and (2) have been formulated in Refs. 5 and 6. In the Born-Oppenheimer approximation it is convenient to characterize the initial quasimolecular term not only by the real part $U_i(R)$, but also by the imaginary contribution $\Gamma(R)$, which is equal to the ionization rate of the quasimolecule A*-B*, when the interatomic distance is R. The probability $P(\rho, \varepsilon)$ that an electron of energy

$$\varepsilon(R)=\varepsilon_{\infty}+U_i(R)-U_f(R), \tag{3}$$

(U_f is the final term of the molecular ion) will be emitted in a collision at the impact parameter ρ of atoms with energy E_i is given (in atomic units) by

$$P(\rho,\varepsilon)=\sum_{s}p(\rho,\varepsilon)\left|\frac{d\varepsilon}{dR_s}\right|^{-1}, \tag{4}$$

where

$$p(\rho,\varepsilon)=\frac{\Gamma(R)}{v_\rho}2\,\mathrm{ch}\left(\int\limits_{R_t}^{R}\frac{\Gamma(R')}{v_\rho}dR'\right)\exp\left(-\int\limits_{R_t}^{\infty}\frac{\Gamma(R')}{v_\rho}dR'\right),$$
$$v_\rho=v_\infty\left(1-\frac{U_i(R)}{E_i}-\frac{\rho^2}{R^2}\right)^{1/2}, \tag{5}$$

R_t is the coordinate of the turning point, and v_∞ is the relative velocity of the atoms at infinity. The potential energies in Eqs. (3) and (5) are taken as zero at infinity: $U_{i,f}(R\rightarrow\infty)=0$. The summation in (4) takes account of the fact that the function $R(\varepsilon)$ is not single valued: one extremum of the difference function $\Delta U = U_i - U_f$ will usually in-

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clude two terms of the sum. To avoid complications associated with the multivaluedness of $\varepsilon(R)$ it is convenient to calculate the spectrum as a function of R,

$$\sigma(R)=2\pi\int_{0}^{\rho_{max}} P(\rho,R)\rho\,d\rho, \tag{6}$$

and then to calculate the desired spectrum $\sigma(\varepsilon)$ from the simple formula

$$\sigma(\varepsilon)=\sum_{s}\sigma(R_s)\left|\frac{d\varepsilon}{dR_s}\right|^{-1}. \tag{7}$$

A difficulty arises in calculating the overlap of the spectra: account must be taken of the quasistationary states produced by electron transitions in the effective potential U_f^{eff}. As was noted in Ref. 7, there are two formulations of the problem that are reasonable, depending on the experimental conditions: the "classical" formulation, in which the centrifugal barrier is assumed to be absolutely impenetrable,[1)] and the "quantum" formulation, in which the centrifugal barrier is assumed to be fully penetrable.

If an electron is emitted at the internuclear distance R the kinetic energies of the atoms at large distances will be related by the formula

$$E_f(R)=E_i-U_i(R)+U_f(R). \tag{8}$$

In the quantum formulation of the problem, therefore, the maximum value of R at which an electron transition results in AI will be determined by the condition $E_f(R)=0$ or

$$E_i=U_i(R_g)-U_f(R_g) \tag{9}$$

and will be independent of the angular momentum l (see Fig. 1). Hence in this approximation the AI and PI spectra will not overlap, and to determine q it will be sufficient to separate the $\sigma(R)$ spectrum at $R=R_g$. The value of Q determined in this manner will be a lower bound to the true value.

In the classical formulation for motion with a given angular momentum l, AI will take place when $R<R_{\text{cl}}(l)$, where $R_{\text{cl}}(l)$ is determined from the condition $E_f=\max(U_f^{\text{eff}})$, or

$$E_i=U_i(R_{cl})-U_f(R_{cl})+U_f(R_{ext})+\frac{l(l+1)}{2\mu(R_{ext})^2}, \tag{10}$$

where R_{ext} is the coordinate of the maximum of the effective potential energy of the ionic term (see Fig. 1) and μ is the reduced mass of the colliding atoms. Condition (10) depends on l, so that AI and PI spectra will overlap at electron energies corresponding to $R_g<R<\max(\text{R}_{\text{cl}})$. For high initial collision energies, $\max(R_{\text{cl}})$ is determined from Eq. (10) with $R_{\text{ext}}=R_{\text{cl}}$ and is obviously equal to the coordinate of the turning point for motion in the effective potentials $U_{i,f}$; hence $\max(l)$, the largest value of l for which an electron transition still results in AI, can be determined from the equation

$$E_i=U_i(R_{cl})+l(l+1)/2\mu R_{cl}^2. \tag{11}$$

When $l>\max(l)$ we get PI. As E_i decreases, the number of partial waves that must be taken into account in calculating the total ionization cross section decreases as $l_{0r}=\rho_{0r}(2\mu E_i)^{1/2}$, while $\max(l)$ as determined from Eq. (11) depends only weakly on E_i since R_{cl} lies in the repulsive region of the potential U_i. From the equation $l_{0r}=\max(l)$ one can find the value $E_i=E_0$ beyond which $\max(R_{\text{cl}})$ is determined from Eq. (10) in which l_{0r} is substituted for l, l_{0r} being the angular momentum at which orbiting sets in for motion with the energy E_i in the potential U_i. By assuming for simplicity that $\max(R_{\text{cl}})\ll R_{\text{ext}}=R_{ie}$ and using the approximation $U_i\approx -D_i(R_{ie}/R)^6$ in the attractive region, we can obtain the esitmate[2)] $E_0\lesssim 0.4D_i$. In collisions with $E\ll E_0$, AI takes place for all $l\ll l_{0r}$. Then by separating the $\sigma(R)$ spectrum into two parts at $R=\max(R_{\text{cl}})$ we obtain the value of Q in the classical approach, and this value yields an upper estimate of the true value of Q.

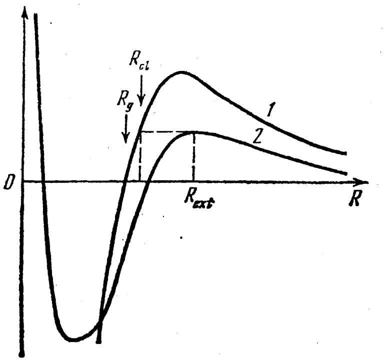

FIG. 1. Kinetic energy E_f of the relative motion of the atom and ion (Eq. (8)) at infinite distance between them vs the distance at which the electron was emitted (curve 1) and the effective potential energy U_f^{eff} for the interaction between the atom and ion (curve 2); R_g and R_{cl} are the maximal distances (determined from Eqs. (9) and (10)) at which the electron transition leads to AI.

We note that in the limit $E_i\to 0$ only s scattering is possible in the initial potential, so that $R_g=R_{\text{cl}}$ and the values of Q calculated in the two approximations should be equal to one another. This suggests that when there is a deep potential well in the term of the initial state, the values of Q calculated from the spectrum in the quantum and classical approximations should be close to one another and to the true value of Q. Actually, if the approximation $U_f\approx -D_f(R_{fe}/R)^4$ is used, the maximum width of the region in which the AI and PI spectra overlap will be

$$\Delta\varepsilon=0.9\left(\frac{R_{ie}D_i^{1/6}}{R_{fe}D_f^{1/4}}\right)^4 E_i^{4/3}. \tag{12}$$

Values of $\Delta\varepsilon$ for symmetric collisions of excited noble gas atoms at $E_i=0.026$ eV are presented in Table I; in some cases use has been made of the similarity between metastable noble gas atoms and alkali metal atoms. The small values of $\Delta\varepsilon$ as compared with the total width of the spectrum provide grounds for assuming that the estimates of Q obtained in the classical and quantum approaches will be close to one another for collisions at thermal energies. In that case the effect of the quasistationary states that arise in connection with the

TABLE I.
Term parameters for the initial (R_{ie}, D_i) and final (R_{fe}, D_f) states and the overlap $\Delta\varepsilon$ of the associative- and Penning-ionization spectra for $E_i = 0.026$ eV.

Colliding atoms	R_{ie}, a_0	D_i, eV	R_{fe}, a_0	D_f, eV	$\Delta\varepsilon$, eV, Eq. (12)
$He(2^3S) - He(2^3S)$	6.5 [4]	0.6 [4]	2 [4]	2.5 [4]	0.22
$Ne(^3P_2) - Ne(^3P_2)$	5.9 [8]	0.75 [8]	3.2 [10]	1.17 [10]	0.06
$Ar(^3P_2) - Ar(^3P_2)$	7.4 [9]	0.52 [9]	4.8 [8]	1.23 [8]	0.02
$Kr(^3P_2) - Kr(^3P_2)$	7.8 [9]	0.49 [9]	5.4 [8]	1.15 [8]	0.02
$Xe(^3P_2) - Xe(^3P_2)$	8.3 [9]	0.45 [9]	6.3 [8]	1 [8]	0.01

centrifugal barrier on the production of molecular ions may be neglected.

Let us consider the $He(2^3S)$–$He(2^2S)$ reaction in more detail. The AI and PI electron spectra of this reaction, averaged over the Maxwell distribution of the atoms at $T = 300$ K, are presented in Fig. 2. In the calculation we used data on the terms and the autoionization width from Ref. 4.

As is evident from Fig. 2, one result of averaging over the velocities is that, according to the quantum calculation, the AI spectrum, which arises from transitions in the small-R region, overlaps the PI spectrum. The low-energy part of the AI spectrum with energies $\varepsilon \gtrsim \varepsilon_\infty$ is associated with transitions incident to collisions of atoms from only the initial part of the Maxwell distribution with $E_i \approx 0$. The Penning electrons that fall into this region of the spectrum, on the other hand, are formed in collisions of fast atoms. The considerable additional broadening of the AI spectrum in the classical calculation is due to collisions of fast atoms with large values of l. In the classical calculation, the high-energy part of the PI spectrum with $\varepsilon \gtrsim \varepsilon_\infty$ is formed by collisions with small l, so the contribution of such collisions to the PI spectrum is minor. As is evident from Fig. 2, the overlap of the spectra is in good agreement with the estimate given by Eq. (12) provided allowance is made for the additional broadening due to averaging over the velocity distribution (of the order of kT).

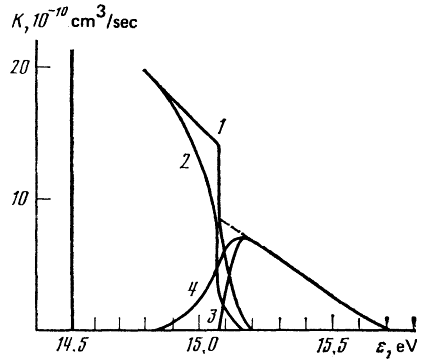

FIG. 2. Electron spectra produced in PI and AI averaged over the velocity distribution of the atoms ($T = 300$ K) and two model calculations. The vertical line at $\varepsilon = 14.5$ eV marks the position of the maximum. 1—The PI spectrum according to the quantum calculation; 2—PI spectrum, classical calculation; 3—AI spectrum, quantum calculation; 4—AI spectrum, classical calculation. The dashed curve is the sum of the calculated spectra; it coincides with curve 1 when $\varepsilon < \varepsilon_\infty$, and with curve 3, when $\varepsilon > 15.2$ eV.

Using the calculated spectra we find that the quantum calculation yields $Q = 10\%$, and the classical calculation, $Q = 14\%$. Therefore, if we have an experimental spectrum obtained, for example, by Penning electron spectroscopy, we can obtain a reliable estimate of Q by separating the spectrum into two parts of $\varepsilon = \varepsilon_\infty + kT$.

We note that this apparently natural separation of the spectrum into two parts is in conflict with the results of Ref. 4, in which Q was also calculated for the $He(2^3S)$–$He(2^3S)$ reaction. According to the results of Ref. 4, $Q \gg 80\%$ in the thermal-energy region; this would indicate that the PI and AI spectra overlap strongly in the classical calculation and that the complete experimental electron spectrum could not be used to determine Q. However, an examination has shown that an error was made in Ref. 4 in calculating Q.[3)]

Among the features of the complete calculated spectrum (the dashed curve in Fig. 2) we note the peak at $\varepsilon = \varepsilon_{ext} = 14.5$ eV, which is associated with an extremum of the difference between the potential curves for the initial and final states.

§3. EXPERIMENTAL TECHNIQUE AND RESULTS

The relation between the electron spectrum $K(\varepsilon)$ arising from reactions (1) and (2) and the velocity distribution function $f(\varepsilon)$ of the fast electrons in the afterglow plasma in the energy region $\varepsilon \gg kT_e$ (T_e is the temperature of the basic Maxwellian group of electrons) was discussed in Ref. 12. It was shown that under conditions in which the fast electrons are lost at the wall of the discharge tube before losing their energy in the bulk of the plasma, the distribution function has the same form, except for a constant factor, as the electron spectrum.

The method of investigating electron spectra from reactions on the basis of measurements of the distribution function $f(\varepsilon)$ in the afterglow plasma in the region of energies ε in which fast electrons appear may be called plasma electron spectroscopy (PLES).

In the present work we used a pulse method, chopping the probe current, to measure the electron velocity distribution function.[13,14] In such measurements the plasma itself serves both as the source of excited particles and as the collision chamber, while an electric probe is used as the analyzer. The comparatively high pressure of the investigated gas in the plasma (≈ 0.1–5 Torr) makes it possible to achieve high concentrations of excited metastable atoms ($\approx 10^{10}$–10^{12} cm^{-3}) in the active phase of the discharge, while as a result of capture of radiation and mixing processes there will be excited atoms at the above concentrations in the investigated stage of the afterglow, not only in metastable states, but also in radiative states associated with them by mixing processes. Two-particle reactions involving excited atoms in various states were selected using the PLES method, either directly from their electron spectra (provided the spectra are separated) or from the change in the intensity of the electron spectrum when the relative concentrations of various excited states are altered.

The limiting resolving power of the PLES method is determined by the temperature of the atoms and amounts to

≈ 0.03 eV if no special measures are taken to cool the plasma. The resolving power could be substantially improved by performing the experiments in a cryogenic plasma.[15,16] To obtain an absolute calibration of the energy scale we used the peak in the electron spectrum that arises from collisions of the second kind between slow electrons and excited atoms in a known state. Analysis showed that such calibration ensures an accuracy of ± 0.1 eV.

The measurements were made in a tube 36 mm in diameter using a movable probe 30 mm long and 0.044 mm in radius; the temperature of the atoms was $T = 300$ K. The PLES method was used primarily to determine Q from the observed spectra for the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^3S)$ and $\mathrm{He}(2^3S)$–$\mathrm{He}(2^1S)$ reactions. A typical result of an experiment after eliminating instrumental broadening due to the method of measurement[17] is presented in Fig. 3. The peak at 14.3 eV is due to the reaction with triplet atoms, and that at 15.4 eV, to the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^1S)$ reaction. In this case the energy resolution was ≈ 0.2 eV. Processes involving metastable atoms in different states were separated as follows. After the end of the active phase of the discharge in the plasma, the deexcitation of singlet metastable atoms by electron impact with their conversion to the triplet state takes place with high efficiency. In the late afterglow at delays of the order of hundreds of microseconds, therefore, under our conditions the concentration of $\mathrm{He}(2^1S)$ atoms turns out to be negligibly small as compared with the $\mathrm{He}(2^3S)$ concentration, and this made it possible separately to obtain the electron spectrum for the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^3S)$ reaction. This, in turn, made is possible to separate the electron spectrum for the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^1S)$ reaction from the total spectrum recorded in the early afterglow.

In accordance with the conclusion drawn from the theoretical treatment, the electron spectrum was separated into two parts corresponding to PI and AI at the energy $\varepsilon = \varepsilon_\infty + kT$, where $\varepsilon_\infty = 15.05$ eV for the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^3S)$ reaction and $\varepsilon_\infty = 15.85$ eV for the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^1S)$ reaction. It was found that the relative number of molecular ions from the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^3S)$ reaction was $(7 \pm 4)\%$; this agrees with the result obtained in Ref. 2 using merging beams and with the results of our calculation. The yield of molecular ions from the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^1S)$ reaction was determined for the first time: $Q = (16 \pm 6)\%$.

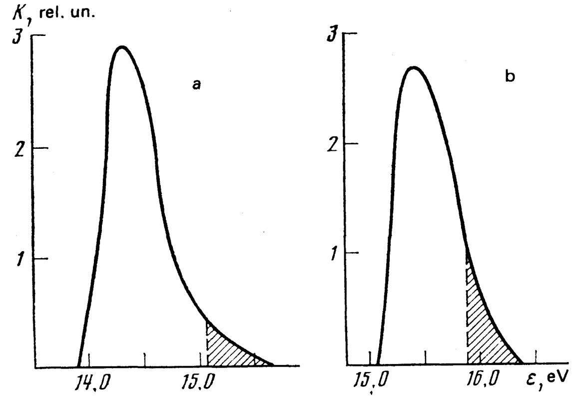

FIG. 3. Experimental electron spectra for the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^3S)$ (a) and $\mathrm{He}(2^3S)$–$\mathrm{He}(2^1S)$ (b) reactions. The hatched regions correspond to electrons emitted in AI processes.

Similar experiments were made with neon, but the sensitivity of the aparatus was such that in this case only an estimate could be made: the yield of molecular ions from the $\mathrm{Ne}(2p^53s^3P_2)$–$\mathrm{Ne}(2p^53s^3P_2)$ reaction was below 25%.

The fact that the width of the observed electron spectrum (≈ 0.4 eV) due to AI in the reactions

$$\mathrm{He}(2^3S) - \mathrm{He}(2^3S), \qquad \mathrm{He}(2^3S) - \mathrm{He}(2^1S),$$

is substantially smaller than the dissociation energy of the He_2^+ molecular ion (2.47 eV) indicates that slow collisions of excited helium atoms lead to the production of He_2^+ ions in highly excited vibrational states. This conclusion also holds for other pairs of noble gas atoms since the high excitation of the vibrational states is essentially due to the substantial difference between the equilibrium separations for the terms of the initial and final states (see Table I).

The shift of the peaks of the calculated (Fig. 2) and experimental (Fig. 3) curves for the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^3S)$ reaction should also be noted. If we assume that the equilibrium distance on the potential curve for the quasimolecule He_2^* lies on a slowly varying portion of the potential curve for the molecular ion He_2^+, we can obtain the dissociation energy for the quasimolecule $\mathrm{He}(2^3S)$–$\mathrm{He}(2^3S)$. This dissociation energy turns out to be 0.9 ± 0.2 eV, while according to the calculation of Ref. 4 it is 0.6 eV. The dissociation energy of the $\mathrm{He}(2^3S)$–$\mathrm{He}(2^1S)$ quasimolecule obtained under the same assumptions is 0.6 ± 0.2 eV.

§4. CONCLUSION

The study of the electron spectra produced in collisions of excited noble gas atoms presented here has established that the overlap of the AI and PI spectra can be neglected and the relative number of molecular ions produced can be determined from the observed spectrum in a simple manner, provided the collision energy is small as compared with the dissociation energy of the initial term. A new experimental method for investigating electron spectra which is promising for the investigation of two-body collisions of excited atoms has been proposed. The study of the spectra from the

$$\mathrm{He}(2^3S) - \mathrm{He}(2^3S), \qquad \mathrm{He}(2^3S) - \mathrm{He}(2^1S),$$
$$\mathrm{Ne}(^3P_2) - \mathrm{Ne}(^3P_2)$$

reactions makes it possible to conclude that comparatively few molecular ions are produced in these reactions ($Q < 25\%$). The molecular ions produced in two-body collisions of excited noble gas atoms are found to be in highly excited vibrational states.

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[1] This formulation of the problem is of interest, in particular, for the case of a plasma having large vibrational relaxation constants.

[2] A model for processes (1) and (2) was proposed in Ref. 7, according to

which ionization takes place only at the turning point for the motion of the atoms. That model yields $Q = 100\%$ when $E \leqslant E_0$.

[3)] Doubts concerning the calculations of Q in Ref. 4 are expressed in Ref. 11 in connection with comparing the calculated Q values with the experimental data.

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