Excitation of metastable states of inert-gas atoms by electron impact in the near-threshold energy region

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An experimental setup is described for a comprehensive investigation of the energy and angular dependences of the formation of metastable particles by a method in which a gasdynamic molecular beam intersects a monoenergetic electron beam, and results of excitation of the metastable states of inert-gas atoms near the threshold are reported. For helium, neon, and argon atoms, a splitting of the maximum of the angular dependence of the produced metastable particles is observed, and it is suggested that it is due to spatial separation of the atoms in two metastable states. The angular distributions of the metastable particles are shown to depend on the mass of the atom and on the energy of the electron. A resonant structure is observed in the energy dependences of the cross sections for the excitation of atoms into metastable states at various detection angles (observation angles). This structure is due to the contribution of the short-lived states of the negative ion. Special observation angles are found, in which the resonances are most clearly pronounced.

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Interest in the study of resonance effects in slow electron–atom and electron–molecule interactions has increased in recent years. One such effect is due to capture of an incident electron with formation of short-lived states of a negative ion, followed by the decay of the ion into an atom in the ground or an excited state and into an electron. The experimental observation and detailed study of these resonances can be effected by several methods. These include experiments on the study of the singularities in the energy dependences of a current of monoenergetic electrons as they pass through a layer of a relatively dense gas (transmission experiment); the determination of the angular and energy distributions of electrons scattered by a beam of atoms, as well as of the energy dependences of individual lines in the loss spectra at fixed scattering angles; and the study of the structure on the optical excitation functions of the spectral lines.

One of the relatively new and promising methods of experimentally investigating resonance effects is the study of the structure on the metastable-state excitation functions (MSEF).[1] This method has a number of undisputable advantages over those listed above. Besides the possibility of obtaining direct information on the shapes and widths of the observed resonances, it has extremely high sensitivity and makes it possible to determine the energy positions of the resonances and identify the negative-ion level to which the resonances belong.[2,3] This method is particularly useful when the energy dependences of the formation of metastable states as well as the angular distributions of the produced metastable particles are investigated in one experiment.[4]

Some experimental material concerning the excitation of metastable states of inert-gas atoms has by now been accumulated. The most complete information is for the helium atom, for which measurements were made for both relative[5,6] and absolute[7,8] excitation cross sections of the 2^3S_1 and 2^1S_0 states. Much less data exist on the excitation of $^3P_{2,0}$ states of other inert gases, for which only the relative energy dependences of the total excitation cross sections were measured in practice.[1,2,9] With the exception of the hydrogen atom,[10] there are likewise no data on the angular distributions of the metastable particles. To be sure, for all the apparent scarcity of experimental data, there is at present detailed information on the resonant structures of the total cross section for the excitation of the metastable states of all the inert-gas atoms,[2,3] and most observed resonances have been classified and identified.[2,11]

We describe in this paper an experimental setup for a comprehensive investigation of the energy and angular dependences of the production of metastable particles excited by monoenergetic electrons, and present new results on the excitation of metastable states of helium, neon, argon, krypton, and xenon atoms near threshold.

1. EXPERIMENT

The kinematic picture of the process of formation of metastable states in slow electron-atom collisions is quite simple.[12] A distinguishing feature of our system of atom and electron beams that interact at a right angle is that both partners move prior to the collision and the target cannot be regarded as at rest. Because of transfer of momentum from the incident electrons to the neutral atoms, the latter experience a recoil that leads to a displacement of their initial trajectory by an angle θ, which we call the observation angle (see Fig. 1), and to a change in the angular distribution of the produced metastable particles compared with the distribution of the primary atomic beam. The angles θ depend on a number of factors: the geometry of the beams, the rates of the colliding particles, the excitation potentials, and the masses of the target atoms. These angles can therefore serve also as an important parameter for the description of the excitation process.

The experiments were carried out with a setup whose block diagram is shown in Fig. 1. An intense beam of inert-gas atoms was produced by the gasdynamic molecular-beam source of the Uzhgorod State

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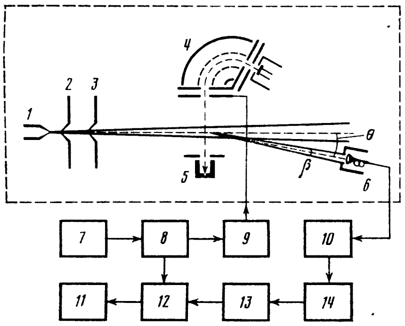

FIG. 1. Diagram of experimental setup and of registration system. 1, 2, 3) Nozzle, skimmer, and collimator; 4) 127-deg cylindrical electrostatic monochromator for electrons; 5) electron receiver; 6) metastable-particle detector (channel electron multiplier); 7) pulse generator; 8) frequency divider; 9) source of accelerating step voltage; 10) preamplifier; 11) number printout unit; 12) multichannel pulse-height analyzer; 13) pulse shaper; 14) amplifier-discriminator.

University[13] (nozzle 1 and slits 2 and 3 in Fig. 1), and crossed at right angle a monoenergetic electron beam shaped by a 127-degree cylindrical electrostatic energy analyzer. The metastable atoms produced as a result of the beam interaction were detected in the beam-intersection plane by a channel electron multiplier 6, located on a movable platform rotating about a center coinciding with the beam intersection point. The high intensity of the atom beam (~10^{15} $cm^{-2} \cdot sec^{-1}$), the negligible energy scatter of the atoms (< 0.1 eV), and the small spatial divergence (not more than one degree), as well as the good monokinetic character of the exciting electrons ($\Delta E = 0.08$ eV at a current ~10^{-8} A) have enabled us to investigate in detail both the angular distributions of the metastable particles and the energy dependences of the excitation cross sections at various observation angles. The angular resolution β of the metastable-particle detector (see Fig. 1) was 0.1°, and the angle θ of rotation of the detector relative to the direction of the neutral beam (see Fig. 1) could range from −10 to +30° and could be set accurate to ±0.2°. The detector null angle was determined with a laser or with a mercury lamp, whose radiation passed through the shaping slits of the gasdynamic molecular-beam source and was incident on the detector. The accuracy with which the null angle is determined was ±0.5° when a laser was used and ±0.15° when ultraviolet radiation of a mercury lamp was used. We have taken special measures to prevent charged and highly excited particles from entering the channel electron multiplier.

The system used to register the metastable particles included (Fig. 1) the channel-electron multiplier 6, a preamplifier 10, an amplifier-discriminator 14, a shaper 13, and an AI-148-2 multichannel pulse-height analyzer 12 equipped with a time analysis device. The useful signal was accumulated in the memory of the pulse-height analyzer over a long time interval by multiple passage through the chosen electron-energy interval. The memory channels of the analyzer and of the step-voltage source that set the value of the electron energy in the collision region in steps of 0.02–0.04 eV, by discretely varying the accelerating voltage of the electron monochromator 4, were scanned with rectangular pulses of a specified frequency (~ 1 Hz) and duration (~10 μsec) from a pulse generator 7 and a frequency divider 8. The accumulated signal from the multichannel pulse-height analyzer was fed to a printout unit 11.

TABLE I.

Atom	Intensity, 10^{15} at·cm^{-2} sec^{-1}	Energy, eV	Mono-kineticity, eV	Diver-gence, deg	Excitation potential of metastable states, eV	Angle range, deg
He	46	0.12	0.06	1.0	19.80; 20.62	0-25
Ne	4.9	0.37	0.07	1.0	16.52; 16.60	0-10
Ar	1.6	0.08	0.02	1.0	11.55; 11.65	0-10
Kr	1.0	0.07	0.01	0.9	9.91; 10.56	0-6
Xe	0.3	0.06	0.015	0.9	8.32	0-6

The measurement procedure consisted of initially determining the range of angles θ for each species of atoms. Next, the excitation functions of the metastable states of the investigated atoms were measured at selected fixed angles θ. We note that the excitation functions obtained in this manner are the energy dependences of the cross sections for the production of metastable states at a fixed observation angle θ, and integration over all the angles θ makes it possible to obtain the total cross section of the process. Naturally, to obtain the absolute values of the cross sections it is necessary to take into account the efficiency of registration of the metastable atom with the aid of the channel electron amplifier, which is different for each atom species as well as for each concrete state.[1)] The results that follow pertain to the summary formation of helium atoms in the triplet and singlet states, and of other inert-gas atoms in the states $^5P_2$ and $^3P_0$ states, since the separation of these states entails considerable difficulties.

The main parameters of the apparatus and the conditions under which the experiments were performed are given in Table I. The neutral-atom beams were generated for each gas in the optimal regime.[13] The time of recording one angular distribution reached 1 hr, and that of oneexcitation function lasted from one to five hours, depending on the magnitude of the useful signal. This time was chosen from the condition for accumulation of enough counts in each analyzer channel to keep their statistical scatter at the maximum of the excitation function lower than 3–5%. The pressure in the collision chamber was maintained during the entire experiment in the range (5-9) × 10^{-7} Torr.

2. EXPERIMENTAL RESULTS AND DISCUSSION

For all the inert-gas atoms, we measured the metastable-state excitation functions from the threshold of the process to the first ionization potential. As already noted above, the first stage in the work was measurement of the angular distributions of the metastable particles at a fixed electron energy. To this end, a definite value of the accelerating voltage was set, and the detector of the metastable atoms (the channel electron multiplier) was rotated in the plane

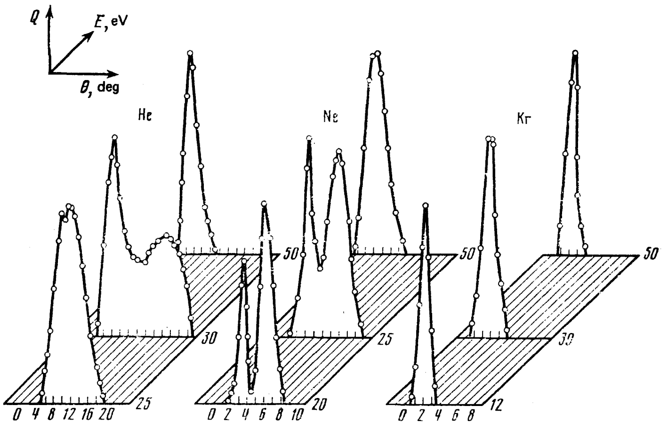

FIG. 2. Angular distributions of metastable atoms of helium, neon, krypton, at various bombarding-electron energies.

of intersection of the beams in the angle range from −10 to +30° in steps of 0.2–1°. The energy dependences were plotted in the automatic regime. The experimental multiple-state excitation-function data was reduced with a computer with graphic display.

The results of our measurements are shown in Figs. 2 and 3. Figure 2 illustrates the angular distribution of the metastable atoms of helium, neon, and krypton. An analysis of these distributions shows that they depend both on the nature of the atom and on the energy of the bombarding electrons. Even though the beams were collimated in approximately the same fashion, the width of the distribution curves at the base depends strongly on the atomic weight. Large difference occur also in the angle θ corresponding to the maximum of the distribution function of the metastable particles. As seen from Fig. 2, for helium and neon one observes a distinct splitting of the principal maximum at relatively low electron energies. This is apparently due to the spatial separation of the atoms produced in two metastable states as a result of the difference between the momentum transferred by the bombarding electrons, a fact most strongly manifest in the energy region close to the threshold. With increasing electron energy, the distribution curve becomes narrower, the maximum tends to the zero angle, and no splitting of the maximum is observed.

The energy dependences of the cross sections for the excitation of the metastable states of the helium, neon, argon, xenon, and krypton atoms were measured by us at fixed angles θ in steps of 1–2°. Figure 3 shows the most typical metastable-state excitation-function curves. We calibrated the electron energy by three independent methods: by comparison of the position of the sharpest peaks on the metastable-state excitation functions of the atoms He, Ne, Ar, Kr, and Xe with the resonances observed by Brunt, King, and Read,[2,3] the positions of which were determined, accurate to ±0.008 eV, from the shift of the current-voltage characteristic of the electron current to the receiver[14] and from the resonant peak on the metastable-state excitation function of the nitrogen molecule at 11.92 eV.[4] The discrepancy between the energy scales determined by these methods did not exceed ±0.02 eV.

As seen from the results, a common regularity of the energy dependences is the presence of a resonant structure and a clear-cut dynamics of its appearance on the metastable-state excitation function with changing angle θ (the numbers over the curves). The good correlation of the observed structure with the energy positions of the known states of the negative ions of the inert gases[15] is evidence that it is due to the contribution of the short-lived states of the negative ions He^-, Ne^-, Ar^-, Kr^-, and Xe^- to the measured cross sections. We note that the energy positions of most observed resonances agree well with the resonances

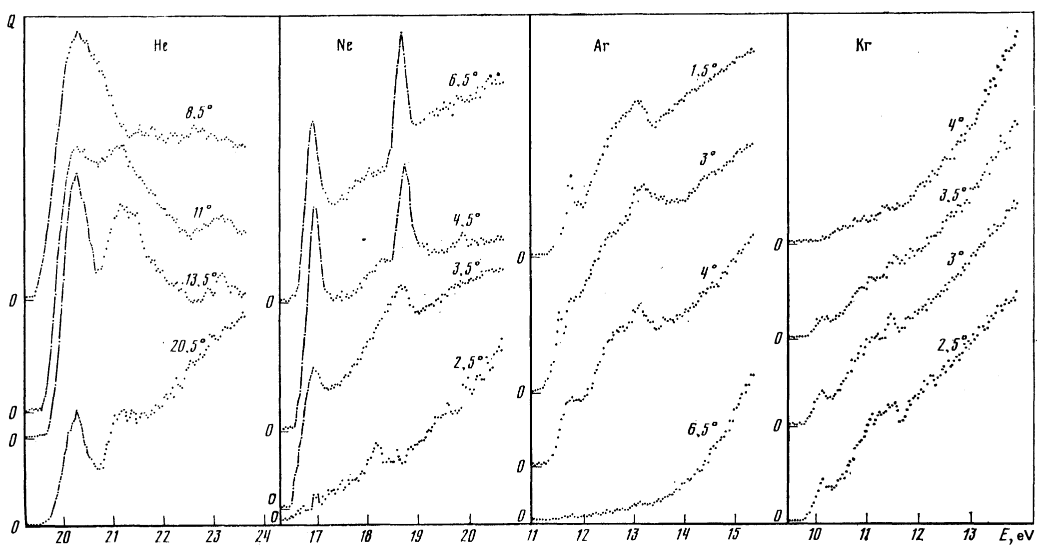

FIG. 3. Energy dependence of the cross sections for the excitation of the metastable states of the atoms He, Ne, Ar, and Kr at various observation angles θ.

observed in the total cross sections for the excitation of the metastable states.[2,3] At the same time, an analysis of the obtained data allows us to conclude that our new procedure is more advantageous because it makes it possible to separate in the definite angles θ, owing to the high angular resolution, those resonances whose contribution to the total cross section of the process is quite insignificant. It should be noted *in passim* that the cross sections measured by Brunt, King, and Read,[2,3] are likewise not total, since the differences between the observation ranges for the light and heavy atoms (see Fig. 2) does not ensure a complete gathering of the metastable atoms that undergo recoil in the course of excitation at a fixed position of the detector.

Helium. The helium-atom metastable state excitation functions measured by us at different angles θ are shown in Fig. 3. This figure illustrates the dynamics of the manifestation of the contribution of an individual resonance in a definite angle. Thus, the maximum at the energy 20.27 eV, which corresponds to formation of a negative ion in 2^2P state, prevails up to an angle $\theta = 11°$, after which a sharply pronounced second resonance is noted at 21.25 eV, due to the contribution of the 3^2S state of the He^- ion, and manifests itself most strongly at an angle 13.5°. Thus, the angles θ equal to 11 and 13.5 as well as to 8.5, 15.5, and 20.5° (see Fig. 3) are special, since they reveal clearly one resonance or another without disturbing the general structural features of the entire curve. At other angles θ (from which, knowing the kinematic relations, it is easy to determine the recoil angles) there are distinguished either a single resonance (7.5°) or several of them (18.5), but whereas the shape and resolution of most observed resonances vary with the observation angle, the energy position of the center of the resonance remains unchanged.

An interesting feature was observed by us near the threshold of excitation of the 2^3S_1 level of helium. As follows from Refs. 3 and 5, the rise of the total excitation cross section curve of the 2^3S_1 level amounts to 0.3 eV, and the first maximum is a broad one. It follows from our results that at certain observation angles (e.g., $\theta = 14.5$ or 18.5°), the metastable state excitation function has near the threshold likewise a broad maximum, whereas at other angles ($\theta = 7.5$, 8.5, 20.5°) the maximum becomes short, and at angles $\theta = 9.5$ and 11° a splitting of this maximum is observed. This is apparently due to the presence of two broad resonances which overlap completely in the total cross section for the excitation of the metastable level, and can be observed only in the differential cross sections for the formation of the metastable state. As for the excitation of the singlet metastable level, its threshold manifests itself distinctly at angles larger than 12.5°.

Neon. A distinct structure and a good resolution of the resonances are typical of the angular and energy dependences of the metastable states of the neon atom (see Figs. 2 and 3). Whereas for helium the doubling of the maxima in the angular distributions are barely noticeable, in the case of neon one observes a good spatial separation of the 3^3P_2 and 3^3P_0 states (see Fig. 2), and the height of the maxima depends significantly on the electron energy. The energy dependences of the production of metastable neon atoms were measured by us in the angle range 2.5–10.5° in steps of 0.5–2° (see Fig. 3). Just as in the case of the helium atom, the dynamics of the manifestation of each resonance singularity on the excitation function depends on the angle θ. We have observed for neon special angles at which only one maximum is observed at 16.91 eV, corresponding to formation of the 3^2P state of the negative neon ion, and a sharply pronounced maximum at 18.67 eV ($\theta = 4.5$ and 6.5°), due to the combined contribution of the 3^1S and 3^1D states.

Other observed structural singularities on the excitation functions correlate well with the data by others with respect to the energy position and the width,[1,2] and agree with the known states of the Ne^- ion. Another interesting fact is that, just for the helium atom, the energy position for the first maximum does not vary with the observation angle.

Argon. Just as in the case of helium and neon atoms, a spatial separation of the 4^2P_2 and 4^3P_0 states as observed for argon (see Fig. 2), but the degree of their resolution is smaller than for the neon atom. This is apparently due to the large mass of the argon atom. Another characteristic fact is that the maxima of the distributions for the different electron energies lie in a narrower range of angles.

The metastable state excitation functions of the argon atom were plotted by us at angles θ from zero to 6.5° (see Fig. 3). In this case, the observation angle is most critical, therefore the energy dependences were taken in steps of 0.5–1.0°. From the obtained family of the curves one can separate the curves measured at the special angles $\theta = 1.5$, 4.5, and 6.5°. At these angles there evolves in succession, in the metastable state excitation functions, a pattern of appearance of resonances corresponding to the formation of 4^3P and 4^1S states of the Ar^- ion with respective energies 11.85 and 13.22 eV. Thus, at an angle $\theta = 1.5°$ the contribution of the 4^3P state of Ar^- to the excitation functions is appreciable, at $\theta = 4°$ this resonance vanishes and the 4^1S state becomes noticeable here, while at $\theta = 6.5°$ the singularities on the metastable-state excitation functions become practically smoothed out.

On the whole, our energy dependences of the production of metastable states of the argon atom have a smaller variation and smaller slope of the metastable-state excitation functions with changing observation angle, although, just as for the objects indicated above, the position of the resonance on the excitation functions at 11.85 eV is independent of the angle θ, and a more gentle rise of the curve is observed with increasing observation angle θ.

Krypton and xenon. The large mass of these elements makes the scattering pictures similar, and consequently the angular distributions of the metastable products are quite similar. By way of example, Fig.

2 shows the distributions of krypton metastable atoms. The range of angles θ is narrow, and no spatial separation of the 5^3P_2 and 5^3P_0 states is observed. The obvious reason is that the detector angular distribution is insufficient for the case of heavy atoms. The energy dependences (see Fig. 3) are characterized by a slow growth of the excitation functions and by the presence of weakly pronounced fine-structure maxima, whose identification presents certain difficulties. This can be apparently attributed to the high degree of screening of the nucleus, which in turn leads, in the case of formation of a negative ion, to a weak coupling of the external electron (and hence to a weak contribution of the resonance to the measured metastable-state excitation function). A common regularity in the metastable-state excitation functions is a weak dependence of the shapes of the curves on the angle θ, but it is not as strongly pronounced as in the case of light atoms.

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[1)]Thus, for helium the ratio of the probabilities of the detection of metastable atoms in the triplet and singlet states is 0.73–0.97 (Ref. 3)

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