Formation of negative carbon ions in charge exchange

I. T. Serenkov, R. N. Il'in, V. A. Oparin, and E. S. Solov'ev

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences
(Submitted September 19, 1974)
Zh. Eksp. Teor. Fiz. **68**, 1686–1692 (May 1975)

The cross sections for negative carbon ion formation by capture of one or two electrons by atoms are positive ions of carbon in Na, Mg, N_2 or O_2 are measured in the 20 to 180 keV energy range. The fraction of negative ions in the excited metastable state in a negative carbon ion beam produced by capture of one or two electrons in Na, Mg, N_2, O_2, Ar or Xe is measured at 30, 100 and 150 keV. An explanation, based on the assumption of conservation of total spin of two electrons simultaneously captured by the C^+ ion, is given for the increase of the fraction of negative ions in the excited metastable state observed in processes involving simultaneous capture of two electrons as compared to processes in which negative carbon ions are produced by capture of a single electron.

PACS numbers: 34.60.

INTRODUCTION

Recent investigations of negative ions have revealed the existence in them of metastable excited states[1-5]. In particular, it was shown in[3] that in electron-capture processes the C^- ion is produced both in the ground state ^4S and in the excited state ^2D, and the state predominantly produced in the experiments was the weakly-bound excited ^2D state, in which the binding energy of the electron was 0.036 eV. It was therefore of interest to investigate the conditions that influence the formation of negative ions in such states in atomic collisions.

In this paper we consider the capture of electrons by carbon atoms and singly-charged positive carbon ions from atoms or molecules of a target, with formation of negative carbon ions. The ion C^- has two experimentally observable states, ^4S and ^2D, with electron binding energies 1.25 and 0.036 eV, respectively, and the fraction of the negative ions in each of these states can be determined by disintegrating the ^2D state in a strong electric field. The processes of the formation of the C^- ion following the capture of electrons were investigated in the energy range from 20 to 165 keV. The targets chosen were Na, Mg, N_2, and O_2, which had different external electron shell structures. The population of the ^2D and ^4S states of C^- were investigated also for Ar and Xe.

DESCRIPTION OF SETUP

Singly-charged positive carbon ions obtained in a high-frequency source operating with CO were accelerated to the required energy and, after passing through a magnetic mass monochromator, were directed to the measuring part of the setup shown in Fig. 1. Chamber I was used to obtain fast carbon atoms, the capture of one electron by which was subsequently investigated. In this case the capacitor C_2 was used to clear the beam of the charged particles. In the investigation of the capture of two electrons ($C^+ \rightarrow C^-$), the gas did not enter into chamber I, and the system of capacitors C_1 and C_2 was used to separate, by parallel displacement, the C^+ beam from the C atoms produced during their flight towards the collision chamber II or III. The cross sections for the capture in Na and Mg vapor were measured in chamber II by using interchangeable heated blocks, and the measurements for the gases O_2 and N_2 were made in chamber III. After passing through the collision chamber, the fast-particle beam was charge-analyzed with a magnetic mass analyzer M. The positive, neutral, and negative components were simultaneously registered in channels 1, 2, and 3 respectively. Channel 4 was used to determine the fraction of negative ions in ^2D states in the C^- beam.

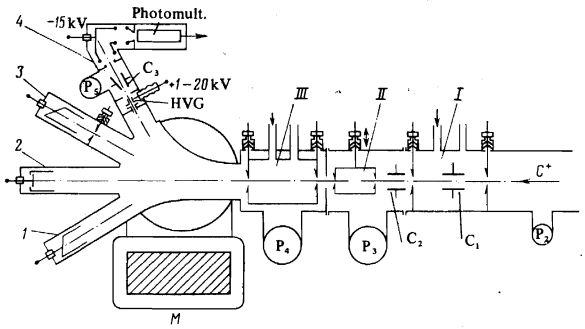

FIG. 1. Diagram of the measuring part of the experimental setup: I–III) target chambers, 1–4) measurement channels, C_1–C_3) deflecting capacitors, P_2–P_5) vacuum pumps, M) mass analyzer, HVG–high-voltage gap.

DETERMINATION OF CROSS SECTIONS

The capture cross sections were determined from the dependence of the current of the C^- ions on the pressure p in the collision chamber. When working with gases, the pressure was measured by three parallel-connected ionization manometers, the scatter of the readings of which did not exceed 15%. The pressure of the Na and Mg vapors was determined from the temperature of the oven, the cross sections being measured during the course of slow cooling of the oven.

When measuring the cross section for the capture of two electrons by a C^+ ion ($\sigma_{1\bar{1}}$), it was necessary to take into account other possible channels of C^- production. An appreciable contribution to the measured current of the C^- ions can be made by the two-step process $C^+ \rightarrow C \rightarrow C^-$, and also by the process $C \rightarrow C^-$, in which the atoms C are produced in the collision of the C^+ ions with the residual gas as they travel from the mass monochromator to the collision chamber. The neutral beam component produced by the residual gas was eliminated by a parallel displacement of the C^+ ion beam by a system of capacitors C_1–C_2 and by a corresponding displacement of the slits in chamber II or III. The contribution of the two-step processes was taken into account by approximating the experimentally-measured dependence of the current of the negative C^- ions on the pressure p

by means of an expression containing terms proportional to the square of the pressure:

$$i_- = i_+[\pi\sigma_{1\bar{1}} + {}^1/_2\pi^2(\sigma_{10}\sigma_{0\bar{1}} + \ldots) + \ldots],$$

where π is the target thickness, $\sigma_{1\bar{1}}$ is the cross section for the capture of two electrons by the C^+ ion, $\sigma_{0\bar{1}}$ is the cross section for the capture of one electron by the C atom, and σ_{10} is the cross section for the capture of one electron by the C^+ ion. The remaining coefficients of π^2 can be neglected because they are small in comparison with $\sigma_{10}\sigma_{0\bar{1}}$.

A possible systematic error in the measurement of the cross sections σ_{10}, $\sigma_{0\bar{1}}$, and $\sigma_{1\bar{1}}$ can be caused by the scattering of the fast-particle beam in the collisions. The strongest influence will be exerted by the scattering in this case in the double-capture reaction $C^+ \rightarrow C^-$. To verify that the entire C^- beam was gathered, the dimensions of the diaphragm ahead of the ion collector in channel 3 was varied from 15 × 15 to 7 × 7 mm. In this case the current of the C^- ions that were produced by 25-keV C^+ ions was decreased by less than 5%. This allows us to assume that the systematic error due to the scattering is much smaller than the mean-squared deviation in the cross-section measurement, which is determined mainly by the error in the measurement of the pressure and is estimated at 20%.

DETERMINATION OF THE POPULATIONS OF THE ^2D AND ^4S TERMS OF THE C^- ION

The fraction of the ^2D states in the beam of C^- ions was determined in channel 4 (Fig. 1). The C^- beam was collimated with a diaphragm of 0.2 mm diameter and passed through the system of electrodes of the high-voltage gap (HVG), between which a field of intensity up to 250 kV/cm was produced, sufficient to ionize all the C^- ions in the ^2D states.[3] The C atoms produced at the output of the high-voltage gap were registered with a detector that counted individual particles. The C^- ions in the ^4S states, which were not affected by the field, were deflected by capacitor C_3. When the voltage was removed from the plates of the capacitor C_3, the detector registered a total particle flux (of the C atoms and the C^- ions in the ^4S states) equal to the flux of C^- ions entering into the high-voltage gap. The fraction of the excited C^- ions in the ^2D states was determined as the ratio of the detector counts with the voltage of capacitor C_3 turned on and off.

The possible systematic errors in the measurement of the populations of the states of the C^- ion are due to the influence of the electron-optical properties of the high-voltage gap on the beam of charged particles, and possibly to the different scattering of the C^- ions when they are produced in the ground and excited states.

The influence of the electron-optical properties of the high-voltage gap on the beam of negative ions passing through it was verified with the O^- ion, which, as is well known[3], has no long-lived states with binding energy ≤ 0.1 eV and is not disintegrated in fields up to 450 kV/cm. Measurements performed at an O^--ion energy 30 keV have shown that if such an error does exist, it is much smaller than the mean-squared deviations in the measurement of the fraction of the ^2D states, which do not exceed 3% in the investigation of the capture of one electron ($C \rightarrow C^-$) and 5% in the capture of two electrons ($C^+ \rightarrow C^-$). The influence of scattering when negative C^- ions are produced in different states was verified by measuring the fraction of the excited state at various points of the beam cross section. The negative carbon ion beam was shifted relative to the entrance diaphragm of the high-voltage gap by slight variation of the field of the mass analyzer M (Fig. 1). Within the scanning interval, which covered more than 90% of the beam intensity, no changes were observed in the fraction of the ^2D states. The experiment was performed at an ion energy 30 keV.

RESULTS AND DISCUSSION

The cross sections obtained by us for the production of negative carbon ions by capture of one electron ($\sigma_{0\bar{1}}$) and two electrons ($\sigma_{1\bar{1}}$), as functions of the energy of the fast particle, are shown in Fig. 2. The figure shows also the cross section σ_{10} for the neutralization of the positive carbon ions.

Comparing the dependences of the cross sections $\sigma_{0\bar{1}}$ and $\sigma_{1\bar{1}}$ on the energy for a given pair of colliding particles, notice should be taken of their similarity, i.e., in a wide interval of the fast-particle energy the rate of these cross sections changes little in comparison with the changes of the cross sections themselves. This singularity can be observed in most cases investigated by Fogel' and co-workers (a review of these studies is contained in Fogel's article[8]), but appears to be

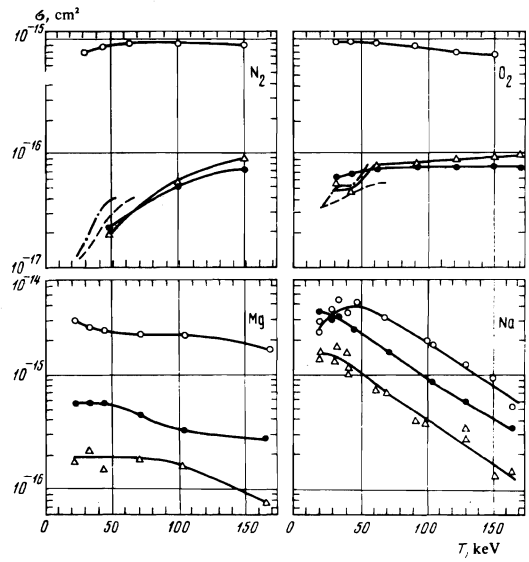

FIG. 2. Cross sections for the capture of electrons from atoms and molecules by carbon atoms and ions as functions of the energy. The type of target is indicated on the diagrams. The symbols for N_2, O_2, and Mg are ○–σ_{10}, ●–$\sigma_{0\bar{1}}$, △–$\sigma_{1\bar{1}}\cdot 10$. The symbols for Na are ○–σ_{10}, ●–$\sigma_{0\bar{1}}\cdot 10$, △–$\sigma_{1\bar{1}}\cdot 100$. Solid curves–results of present study, dashed–results of [6] for $\sigma_{0\bar{1}}$, dash-dot–results of [7] for $\sigma_{1\bar{1}}$.

	Energy of C and C⁺, keV	Targets					
		N_2	O_2	Ar	Xe	Na	Mg
Process C — C⁻							
Measured	30	67±3	55±3	58±3	60±3	66±3	62±3
»	100	63±3	63±3	66±3	64±3	69±3	65±3
»	150	—	—	—	—	68±3	65±3
Calculated		71	71	71	71	71	71
Process C⁺ — C⁻							
Measured	30	73±5	62±5	62±5	70±5	60±5	80±5
»	100	78±5	75±5	82±5	82±5	70±5	90±5
»	150	—	—	—	—	70±5	95±5
Calculated		83	80	83	83	78	100

typical of processes with formation of negative ions in the velocity region where these cross sections are close to maximal.

The table lists the fractions of the ions that are in excited ^2D states as percentages of the total beam of negative carbon ions. For all the investigated pairs, this quantity increases slowly with increasing collision energy and remains practically constant, for a given pair of colliding particles, at energies above 100 keV. In the case of capture of one electron, the fraction of the excited negative ions in the beam is close to the statistical fraction and is practically independent of the type of target particle. If it is recognized that the ^2P states of the C^- ion lie in the continuous spectrum[9, 10], then the relative statistical weight of the ^2D term amounts to 10/14 or 71%. This quantity is the upper limit of the experimental data for the process $C \to C^-$. Upon capture of two electrons (the process $C^+ \to C^-$), the fraction of the negative ions in the excited state depends significantly on the form of the target. For all targets, with the exception of Na, it is higher than in the capture of one electron, and is close to unity for capture in Mg vapor.

It is natural to assume that the main contribution to the cross section for the production of the C^- ions is made by capture of two electrons from the outer shell of the target particle. In the case of magnesium, the outer shell contains two 3s electrons, and their total spin is S = 0. If their total spin remains unchanged when these electrons are captured by a C^+ ion in the ^2P state, then only ^2D-term states of the C^- ion should be observed in the experiment. In our measurements, the fraction of the C^- ions in the ^2D states reaches in this process 0.95, i.e., it differs significantly from the relative statistical weight of the ^2D term. This fact proves, in our opinion, that the reaction follows the scheme $C^+(^2P) + Mg(^1S) \to C^-(^2D) + Mg^{2+}(^1S)$, i.e., it offers evidence that the total spin of the colliding particles is conserved in the reaction. For the two captured electrons, this requires that their spin be conserved. The influence of the conservation of the total spin of two electrons captured in one collision act on the measured capture cross sections was noted by us earlier[11].

We can estimate the influence of the factors noted above, namely the statistical weights of the terms of the isolated C^- ion and the conservation of the total spin of the captured electrons, on the relative population of the terms of the C^- ion. Let us consider the capture of a pair of electrons from the outer shell of a target particle by a C^+ ion, assuming that their total spin is determined by the values of the quantum numbers m_S which they possessed in the target, and that this spin is conserved when this pair of electrons goes over to the C^+ ion. For targets having a singlet or doublet ground-state term, the outer shell with n electrons can be represented as the sum of n_+ electrons with $m_S = 1/2$ and n_- electrons with $m_S = -1/2$. Using simple probability considerations, we can determine the probability of capturing from such a shell two electrons having a summary spin projection $M_S = 0$. Denoting this probability by w_0, we obtain

$$w_0 = 2n_+n_-/n(n-1).$$

In the case of targets having a triplet ground-state term, for example O_2, the probability w_0 was defined as the sum of the quantities calculated for two sets $(n_+ + n_-)$ corresponding to projections of the summary target spin $|M_S| = 1$ and $M_S = 0$, taken with weight factors 2/3 and 1/3. The probabilistic approach to the capture of two electrons from different shells, as for example in the case of sodium, leads directly to $w_0 = 1/2$.

The probability W_0 of observing a total spin for a pair of captured electrons is obtained from the quantities w_0 and $w_1 = 1 - w_0$, recognizing that a unity total spin, S = 1, has three possible projections M_S. Therefore $W_1 = 3w_1/2$ and $W_0 = w_0 - w_1/2$.

If it is recognized that only ^3D states are produced when two electrons with S = 0 are captured by a C^+ ion in the ^2P state (we are considering only the experimentally observable states), and that at S = 1 production of ^4S and ^2D states is possible, and it is assumed that in this case the terms are populated in proportion to their statistical weights, then the fraction of the ^2D states in the beam of C^- ions can be expressed in the form $F = W_0 + W_1 g$, where g is the relative statistical weight of the ^2D term. The values obtained in this manner are listed in the last line of the table. Different refinements, for example allowance for the formation of states of a ^2P term or statistical population of the terms of the residue of the target, lead to close values of F. Comparison of the calculated values of F with the measured ones shows that the calculated data are in good agreement with the experimental ones for collision energies higher than 100 keV ($v > 1.2 \times 10^8$ cm/sec). The largest discrepancy is observed in the case of sodium.

The noted agreement between calculation and experiment enables us to assume that the processes of production of the negative C^- ion in different states at collision velocities exceeding 10^8 cm/sec depend to a considerable degree on the statistical characteristics of the isolated particles, at any rate when the energy consumed in the process greatly exceeds the difference between the energy consumptions for the different channels of the process determined by the final terms of the colliding particles.

The authors are grateful to D. A. Varshalovich for taking part in a discussion of the results.

[1]M. L. Seman and L. M. Branscomb, Phys. Rev. **125**, 1602 (1952).
[2]J. F. Paulson, J. Chem. Phys., **52**, 5491 (1970).
[3]V. A. Oparin, R. N. Il'in, I. T. Serenkov, E. S. Solov'ev, and N. V. Fedorenko, ZhETF Pis. Red. **13**, 351 (1970) [JETP Lett. **13**, 249 (1970)].
[4]V. A. Oparin, R. N. Il'in, I. T. Serenkov, E. S. Solovyov and N. V. Fedorenko, Electronic and Atomic Collisions, Abstracts of papers VII ICPEAC, Amsterdam, 1971, p. 796.
[5]D. Feldman, Zs. Naturforsch., **26a**, 1100 (1971).
[6]Ya. M. Fogel', V. A. Ankudinov, and D. V. Pilipenko, Zh. Eksp. Teor. Fiz. **34**, 579 (1958) [JETP Lett. **7**, 400 (1958)].
[7]Ya. M. Fogel', R. V. Mitin, and A. G. Koval', Zh. Eksp. Teor. Fiz. **31**, 397 (1956) [Sov. Phys.-JETP **4**, 359 (1957)].
[8]Ya. M. Fogel', Usp. Fiz. Nauk **71**, 243 (1960) [Sov. Phys.-Uspekhi **3**, 390 (1960)].
[9]B. L. Moiseiwitsch, Adv. Atom. Molec. Phys., **1**, 61 (1965).
[10]J. Hunt and B. L. Moiseiwitsch, J. Phys. B., **3**, 892 (1970).
[11]R. N. Il'in, V. A. Oparin, I. T. Serenkov E. S. Solovyov and N. V. Fedorenko, Electronic and Atomic Collisions, Abstracts of papers VII ICPEAC, Amsterdam, 1971, p. 793.

Translated by J. G. Adashko
181