Scattering of negative moderate-energy chlorine ions by atoms and molecules

R. N. Il'in, V. I. Sakharov, and I. T. Serenkov

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

(Submitted 20 July 1988)
Zh. Eksp. Teor. Fiz. **95**, 1256–1262 (April 1989)

The differential scattering cross sections have been determined for electron detachment from a Cl^- ion colliding with an argon or sodium atom or with a CO molecule at energies of 45, 100, and 200 keV. A dynamic detachment mechanism acting at high impact parameters was found to play a considerable role. Inelastic scattering of chlorine atoms was observed for the Cl^-–Na pair.

Detachment of an electron from a negative ion, which usually does not have long-lived excited states, represents a transition from the only bound state to a continuous spectrum and therefore represents a suitable topic for experimental and theoretical studies. The interest in such detachment is partly due to the fact that in the case of the H^- ions it is one of the main processes used in injectors suitable for thermonuclear reactors.

The detachment mechanism depends on the collision velocity. For $v \ll v_e$, where v_e is the 'orbital" velocity of a weakly bound electron, in the case of internuclear distances $R < R_0$, where R_0 corresponds to the equality of the potential energies of the quasimolecular systems $(AB)^-$ and $(AB + e)$, a weakly bound electron goes over to a continuous spectrum. This mechanism will be called quasimolecular and it can be described by the complex-potential method[1] or by a zero-radius potential,[2,3] and in the latter case it is possible to allow for the contribution of transitions under the barrier which occur when $R > R_0$.

In the velocity range $v > v_e$ detachment results from the scattering of a weakly bound electron on an atom or molecule in a target. We shall call this the dynamical mechanism: it can be described both quantum-mechanically[4] and by classical models.[5,6] For $v \gg v_e$, the detachment problem can be solved by the Born method.

The velocity range $v \leqslant v_e$ is intermediate between the two other cases. In our earlier paper[6] we analyzed the total cross sections σ_{10}^- for the detachment of an electron from a Cl^- ion and we concluded that both these mechanisms were active simultaneously, and that the contribution of the dynamic mechanism increased with the velocity. The possibility of simultaneous action of both mechanisms in the case of the H^- ions was pointed out also in Ref. 7 reporting work carried out independently and simultaneously with us.

An investigation of the scattering in the course of detachment of an electron from a negative ion provides more detailed (compared with measurements of the total cross sections) information on the process and in particular on the transition from one detachment mechanism to another not only when the velocity is varied, but also when the impact parameter is altered.

We determined the differential scattering cross sections $d\sigma/d\Omega$ of Cl^- ions of energies 45, 100, and 200 keV corresponding to velocities of 0.23, 0.34 and $0.48v_0$, where v_0 is the atomic unit of the velocity. The orbital velocity of a weakly bound electron in a Cl^- ion is $v_e = 0.73v_0$ (Ref. 6). Our targets were the atomic Ar and Na, which we used earlier,[6] as well as CO molecules. These targets were selected because the scattering of the Cl^- ions by the atoms of Ar and Na had been investigated in the range $v \ll v_e$ and reported in Refs. 8 and 9, respectively. It was shown in Ref. 8 that at ion energies $0.75 \leqslant E \leqslant 2$ keV the detachment process is governed by the quasimolecular mechanism, which is the reason (in particular) for the threshold value of the angle. When the ions of Cl^- were scattered by Na in the energy range $0.5 \leqslant E \leqslant 1$ keV (Ref. 9), only the elastic scattering was observed and the inelastic cross section was too small to measure. We demonstrated earlier[6] that in the energy range $20 < E < 200$ keV the cross sections were $\sigma_{10}^- > 1 \times 10^{-16}$ cm^2. They increased with the velocity, in accordance with the dynamical mechanism model developed in Ref. 6.

Collisions of negative ions with some molecules (CO, N_2, CO_2) make it possible to observe a third mechanism of detachment via charge exchange, yielding an unbound resonant state of a molecular negative ion, which then decays. Our determination of the total cross sections for the detachment of an electron from various negative ions colliding with CO molecules indicated that these cross sections have a strong maximum at ion velocities close to the velocities corresponding to a shape resonance ($^2\Pi$) in the scattering of a free electron.[10,11] The recent investigations of the detachment of an electron from the Cl^-, F^-, and H^- ions colliding with molecules[12–15] also demonstrated that charge exchange plays an important role, yielding an unbound discrete state (this was deduced from an analysis of the energy spectra of the atoms and free electrons formed in the detachment process), but there are practically no data on the scattering of atomic particles at velocities close to the resonance value.

EXPERIMENTAL METHOD

Ions were generated using a high-frequency source by admission of carbon tetrachloride vapor into a chamber. Negative ions were formed as a result of charge exchange with suitable positive ions in the output channel of the ion source. Next, a beam of negative ions was focused and accelerated to the required energy. After magnetic analysis and collimation a beam entered a collision chamber filled with a gas or a vapor. The scattered particle beam emerging from this chamber crossed a transit region and reached a fixed detector in the form of a chevron array of two 60-mm microchannel plates. The collimator consisted of two vertical slits 10 μm wide and 0.6 mm high separated by a distance of 80 cm. The length of the collision chamber used in the case of gas targets amounted to 16.5 cm. A sodium vapor target was constructed as described in Ref. 16 and in this case the length

 0038-5646/89/040724-04$04.00

of the collision chamber was 11 cm. The length of the transit region (from the center of the chamber to the detector) was 327 cm. A measuring slit 35 μm wide and 4 cm high was placed in front of the detector. The thickness of the gaseous target used in the cross section measurements was less than or equal to 2×10^{14} cm^{-2} and the thickness of the sodium vapor target was 0.5×10^{14} cm^{-2}, which ensured that the collisions were single.

The angular dependences were obtained by applying steplike voltages to a pair of vertical plane-parallel plates located directly in front of the collision chamber. The signal intensity was measured for each value of the potential difference between the plates. This was a modification of the familiar method used in determination of the beam divergence (see, for example, Ref. 17). The signals from the detector and the step voltages from the plate were picked up by an automatic system controlled by a microcomputer which was linked to a counter and a digital-analog converter. The profile of a beam scattered by the target (representing the dependence of the beam intensity on the deflection angle of the primary beam) was defined as the difference between the profiles obtained at working and residual pressures in the collision chamber, which made it possible to avoid the errors associated with the scattering on slits and in the residual gas.

The angular dependence of the differential cross sections was investigated by subjecting the experimental curves to a reconstruction procedure similar to that described in Ref. 18. This was necessary because ribbon-shaped beams and an extended slit in front of the detector were used in our experiments. The absolute values of the differential cross sections of the detachment process were obtained by calibrating curves normalized to the total cross sections we determined earlier[6]; the absolute elastic scattering cross sections were formed from the geometrical parameters of our experiments. By way of control, we determined the absolute differential cross section for electron detachment in the case of a Cl–Ar pair at an energy of 100 keV and we compared the results with the cross section obtained by the calibration method; the curves disagreed by less than 20%.

RESULTS OF MEASUREMENTS AND DISCUSSION

Cl^-–Ar pair

The measured differential cross sections for the detachment of an electron $\rho_{\bar{1}0}$ and of the elastic scattering $\rho_{\bar{1}\bar{1}}$ of the Cl^- ions are plotted in Fig. 1 using reduced coordinates $\rho = (d\sigma/d\Omega)\theta \sin\theta$ and $\tau = \theta E$ (θ is the scattering angle). For comparison, this figure includes also the results of Ref. 8 for $E = 2$ keV, normalized to the total cross section using the data from Ref. 19. Figure 2 shows the dependence

$$P_d(\tau) = \rho_{\bar{1}0}(\rho_{\bar{1}0}+\rho_{\bar{1}\bar{1}})^{-1}$$

deduced from the experimental data. Using model representations developed in Refs. 8 and 20, we concluded that the function $P_d(\tau)$ approximately described the detachment probability, whereas the relationship between the impact parameter b and the reduced angle τ could be obtained by calculating the elastic scattering on the Cl^-–Ar potential.[8] In this approximation the value $R_0 = 3.39a_0$ (a_0 is the Bohr radius) corresponds to a reduced angle $\tau_0 = 1.26$ keV·deg. The curves in Figs. 1 and 2 show that an increase in the collision velocity increased the contribution made to the total scattering by those processes which occured at values of the parameters considerably exceeding R_0. This was in conflict with the conclusion reached in Ref. 21, viz., that the rise of the total cross section in the range $E > 10$ keV was due to a growing contribution of the detachment via the formation of excited states Cl* and Ar*, because (as shown in Ref. 22) such processes occured at parameters much smaller than R_0.

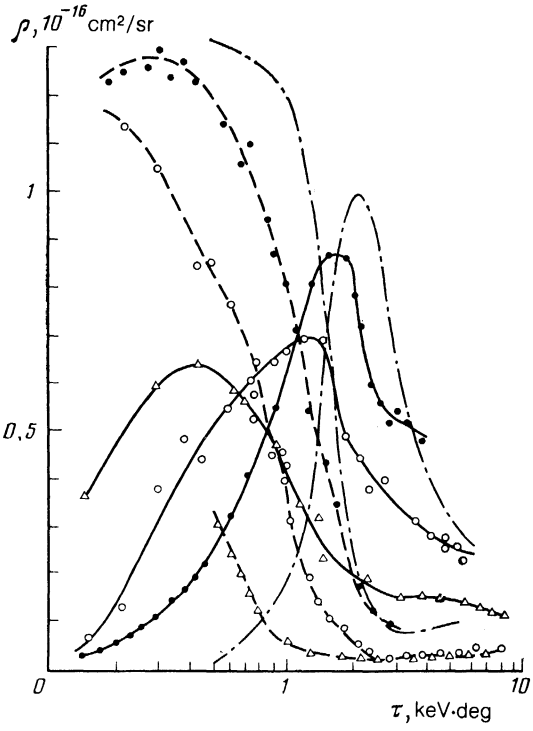

FIG. 1. Differential cross sections of the processes of detachment (continuous curves) and elastic scattering (dashed curves) obtained for the Cl^-–Ar pair at energies $E = 45$ keV (●), 100 keV (○), and 200 keV (△). The chain curves represent the results of Ref. 8 for $E = 2$ keV.

As shown in Ref. 3, some increase in the detachment

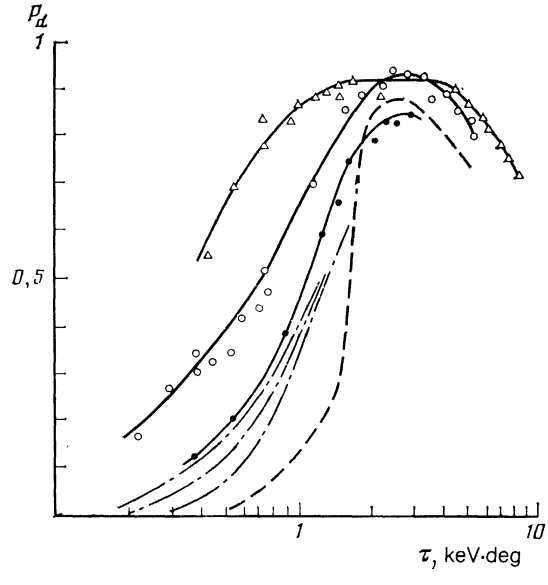

FIG. 2. Electron detachment probabilities for a Cl–Ar pair at energies $E = 45$ keV (●), 100 keV (○), and 200 keV (△). The chain curve represents the calculated probability of subbarrier transitions at energies $E = 45$, 100, and 200 keV (an increase in the probability at a fixed value of τ corresponds to an increase in the energy); the dashed curve represents the results taken from Ref. 8 and obtained at $E = 2$ keV.

probability in the $\tau < \tau_0$ case with increasing velocity can be attributed to an increase inthe contribution of subbarrier transitions. We used a calculation method developed in Ref. 3 to obtain the relevant estimates and we employed parameters determined on the assumption that in the range $\tau < \tau_0$ at $E = 2$ keV the dependence $P_d(\tau)$ was governed by subbarrier transitions. The results of our calculations, also plotted in Fig. 2, demonstrate that a strong increase in the probability of detachment in the range $\tau < \tau_0$ as a function of the velocity cannot be explained simply by an increase in the contribution of subbarrier transitions. Moreover, the contribution of subbarrier transitions cannot account for the experimentally observed monotonic rise of the differential detachment cross section $d\sigma_{\bar{1}0}(\theta)/d\Omega$ as the scattering angle decreases down to $\theta = 0$ [allowing for the resolution amounting to $\Delta\theta = (2\text{–}3) \times 10^{-5}$ rad].

The rise of the detachment probability in the range $\tau < \tau_0$ which we observed and the associated shift of the maxima of the reduced cross sections toward lower reduced angles can be explained by an increase in the contribution of the dynamical detachment process typical of fast collisions. Estimates of the differential detachment cross section as a function of the scattering angle,[7] based on the scattering of a weakly bound electron, confirmed the monotonic rise of the cross section in the limit $\theta \to 0$.

In the range $\tau > \tau_0$ we can see from Fig. 2 that the function $P_d(\tau)$ agrees well (at the energies used in our study) with the corresponding dependence for $E = 2$ keV. Hence, we may conclude that at low impact parameters the quasimolecular detachment mechanism predominates. Therefore, the results of our measurements confirm our earlier conclusion[6] that two detachment mechanisms exist simultaneously in the intermediate range of velocities and a transition takes place from the quasimolecular to the dynamical mechanism as the particle velocity increases. Rough estimates of the contribution of the dynamical mechanism to the total detachment cross section, obtained from the dependence $P_d(\tau)$ (Fig. 2), demonstrate that an increase in the energy from 50 to 200 keV changes the contribution of the dynamic mechanism from 30 to 75 %, which is in satisfactory agreement with the results of a calculation reported in our earlier paper.[6]

Cl^-–Na pair

The results of measurements of the differential cross sections for the detachment of an electron at energies of $E = 45$, 100, and 200 keV and also of the elastic scattering of $E = 45$ keV Cl^- ions are plotted in Fig. 3 using the reduced coordinates. This figure includes the elastic scattering cross section calculated by a semiclassical method described in Ref. 9. The dependence $\rho_{\bar{1}\bar{1}}(\tau)$ reveals clearly a maximum of the rainbow scattering at a calculated position corresponding to a reduced angle of $\tau_R = 0.107$ keV·deg.

The $\rho_{\bar{1}0}(\tau)$ curves have two maxima. The first, lying in the range $\tau > 1$ keV·deg and corresponding to small impact parameters, makes the dominant contribution to the total detachment cross section, which—as shown in Ref. 6—is governed by the dynamical mechanism. Note that the position of the plateau on the calculation dependence $\rho_{\bar{1}\bar{1}}(\tau)$ corresponds to the internuclear distance at which the $3p$ shell of the Cl^- ion overlaps the $2p$ shell of the Na atom.

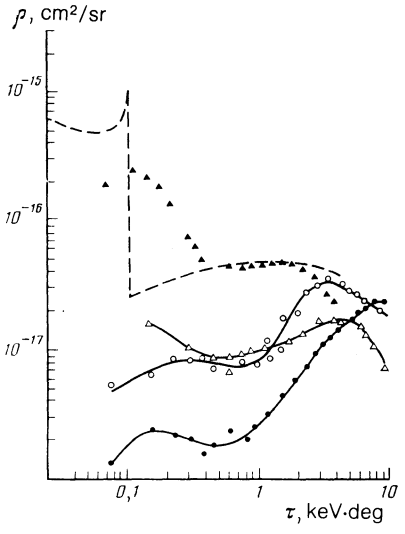

FIG. 3. Differential cross sections of detachment and elastic scattering processes for the Cl^-–Na pair. The continuous curves represent detachment at energies $E = 45$ keV (●), 100 keV (○), and 200 keV (△). The black triangles represent elastic scattering at $E = 45$ keV. The dashed curve is calculated for the elastic scattering by the semiclassical method.

The second maximum, located in the range of reduced angles close to τ_R, is due to rainbow scattering. The appearance of a rainbow in the inelastic interaction case is in this case due to the fact that the loss of an electron for impact parameters close to $5a_0$ i.e., close to the value of R corresponding to the reduced angle τ_R, causes a transition of the system to the ground term (Na^+Cl^-) resulting from the ionic binding and exhibiting a minimum in the above-mentioned range of the internuclear distances.[9]

On the whole, the behavior of the differential detachment cross section for the Cl^-–Na pair exhibit the same characteristics typical of the dynamical mechanism as does the Cl^-–Ar pair: the reduced cross section rises with energy at low reduced angles, the main maximum (which is not associated with the rainbow) shifts toward lower values of τ as the energy increases from 45 to 100 keV, and the differential cross section $d\sigma_{\bar{1}0}(\theta)/d\Omega$ rises in the limit $\theta \to 0$.

Cl^-–CO pair

The results of measurements of the differential detachment cross sections for this pair are presented in Fig. 4. This figure also includes, for the sake of comparison, the dependences $\rho_{\bar{1}0}(\tau)$ for Cl^-–N_2 (148 eV) and F^-–N_2 (1 keV) pairs, which are plotted using the data taken from Refs. 13 and 15, respectively. The similarity of the structures of the Cl^- and F^- ions and also of the properties of the N_2 and CO molecules allows us to use these data in rough comparisons.

The dependence $\rho_{\bar{1}0}(\tau)$ determined in the present study for $E = 45$ keV has two maxima; raising the energy increases the first maximum corresponding to low values of τ, whereas the second maximum located at $\tau \approx 1$ keV·deg falls steeply. The main contribution to the total cross section comes from the first maximum and, as pointed out above, in this range of energies the total cross section is governed by the process of charge exchange creating an unbound state $^2\Pi$ of the CO^- ion. Hence it is natural to assume that the first maximum of the dependence $\rho_{\bar{1}0}(\tau)$ corresponds to charge exchange pro-

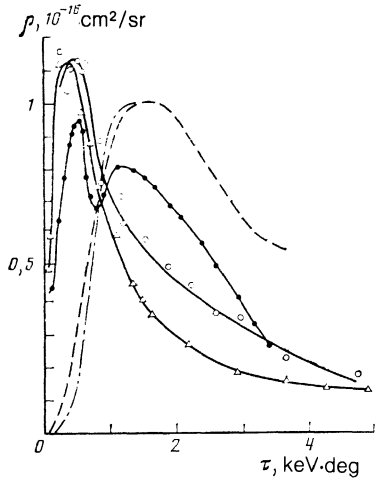

FIG. 4. Differential detachment cross sections for the Cl–CO pair at energies $E = 45$ keV (●), 100 keV (○), and 200 keV (△). The chain curve represents the detachment cross section in arbitrary units for the Cl^-–N_2 pair (148 eV) taken from Ref. 13, whereas the dashed curve is the detachment cross section, in arbitrary units, for the F^-–N_2 pair (1 keV) taken from Ref. 15.

ducing an unbound state at higher values of the impact parameters, whereas the second maximum represents detachment resulting from the crossing of a surface representing the potential energy of the original $Cl^- + CO$ system by a surface representing the boundary of the continuous spectrum ($Cl + CO + e$). This view is supported by the positions of the maxima of the dependences $\rho_{\bar{1}0}(\tau)$ observed for the Cl^-–N_2 and F^-–N_2 pairs at low energies when the quasimolecular detachment mechanism predominates.

The opposite assumption, that the initial term first intersects the boundary of the continuous spectrum and then charge exchange occurs at low parameters when the surfaces of the potential energy of the $Cl^- + CO$ and $Cl + CO^-$ systems intersect, cannot explain the large value of the cross section of this process. Hence, we may conclude that the process of detachment as a result of charge exchange to an unbound discrete resonant state should be regarded as the result of a direct interaction of a weakly bound electron with the target molecule and the description of this process should probably be based on the momentum methods, as was done in Refs. 14 and 23.

We conclude with the following comments. Investigations of the process of scattering of the Cl^- ions, accompanied by the detachment of an electron due to collisions with Ar or Na atoms and CO molecules, show that in the range of velocities $(0.23–0.48)v_0$ the detachment may be due to several simultaneous mechanisms and their relative contribution to the total cross section depend on the collision velocity. In the case of the Cl^-–Ar and Cl^-–CO pairs at velocities close to the lower limit of the range under consideration a considerable role is played by the quasimolecular mechanism which is due to a shift of the initial term to the continuous spectrum. An increase in the velocity in the case of the Cl^-–Ar pair causes the dynamical mechanism to dominate. The latter is characterized by a high probability of detachment at parameters greater than R_0 and an increase in the differential cross section $d\sigma_{\bar{1}0}(\theta)/d\Omega$ the scattering angle θ is reduced down to $\theta \approx 0$. In the case of the Cl^-–Na pair this mechanism is the only one throughout the range of velocities investigated. In the case of the Cl^-–CO pair at velocities of $(0.34–0.48)v_0$ the dominant mechanism involves charge exchange to the unbound state $^2\Pi$ of the CO^- ion. Such charge exchange occurs at parameters exceeding $R_\times$ which is the distance corresponding to the crossing of the $Cl^- + CO$ and $Cl + CO^-$ terms.

Collisions of the Cl^- ions with the Na atoms revealed a rainbow not only for the elastic scattering, but also for the inelastic scattering, which is explained by a similarity of the form of the interaction potential as a function of R for the $Cl^- + Na$ and $Cl + Na$ systems.

The authors are deeply grateful to V. V. Afrosimov for a valuable discussion of this work.

[1]R. L. Champion and L. D. Doverspike, Phys. Rev. A **13**, 609 (1976).
[2]Yu. N. Demkov, Zh. Eksp. Teor. Fiz. **46**, 1126 (1964) [Sov. Phys. JETP **19**, 762 (1964)].
[3]A. Z. Devdariani, Zh. Tekh. Fiz. **43**, 399 (1973) [Sov. Phys. Tech. Phys. **18**, 255 (1973)].
[4]G. B. Lopantseva and O. B. Firsov, Zh. Eksp. Teor. Fiz. **50**, 975 (1966) [Sov. Phys. JETP **23**, 648 (1966)].
[5]D. R. Bates and J. C. G. Walker, Proc. Phys. Soc. London **90**, 333 (1967).
[6]I. T. Serenkov, R. N. Il'in, and V. I. Sakharov, Zh. Eksp. Teor. Fiz. **88**, 417 (1985) [Sov. Phys. JETP **61**, 243 (1985)].
[7]P. E. van der Leeuw, A. Tip, W. Koot, *et al.*, Chem. Phys. **101**, 183 (1986).
[8]C. de Vreugd, R. W. Wijnaendts van Resandt, J. B. Delos, and J. Los, Chem. Phys. **68**, 261 (1982).
[9]C. de Vreugd, R. W. Wijnaendts van Resandt, J. Los, *et al.*, Chem. Phys. **42**, 305 (1979).
[10]I. T. Serenkov, R. N. Il'in, and V. A. Oparin, Zh. Tekh. Fiz. **47**, 860 (1977) [Sov. Phys. Tech. Phys. **22**, 515 (1977)].
[11]R. N. Il'in, V. I. Sakharov, and I. T. Serenkov, Abstracts of Papers presented at Twelfth Intern. Conf. on Physics of Electronic and Atomic Collisions, Gatlinburg, TN, p. 535.
[12]J. S. Risley, Phys. Rev. A **16**, 2346 (1977).
[13]B. K. Annis, S. Datz, R. L. Champion, and L. D. Doverspike, Phys. Rev. Lett. **45**, 1554 (1980).
[14]Vu Ngoc Tuan, V. Esaulov, J. P. Gauyacq, and A. Herzenberg, J. Phys. B **18**, 721 (1985).
[15]Vu Ngoc Tuan, V. A. Esaulov, J. P. Grouard, et al., J. Phys. B **17**, 2897 (1984).
[16]C. J. Anderson, R. J. Girnius, A. M. Howald, and L. W. Anderson, Phys. Rev. A **22**, 822 (1980).
[17]L. L. Ames, Nucl. Instrum. Methods **151**, 363 (1978).
[18]E. P. Gorbunov, Diagn. Plazmy No. 3, 358 (1973).
[19]Yu. F. Bydin and V. M. Dukel'skiĭ, Zh. Eksp. Teor. Fiz. **31**, 569 (1956) [Sov. Phys. JETP **4**, 474 (1957)].
[20]V. Esaulov, D. Dhuicq, and J. P. Gauyacq, J. Phys. B **11**, 1049 (1978).
[21]B. Hird and F. Rahman, J. Phys. B **16**, 3581 (1983).
[22]J. Fayeton, D. Dhuicq, and M. Barat, J. Phys. B **11**, 1267 (1978).
[23]A. K. Kazanskiĭ, Zh. Eksp. Teor. Fiz. **86**, 1995 (1984) [Sov. Phys. JETP **59**, 1160 (1984)].

Translated by A. Tybulewicz