SCATTERING OF HIGHLY EXCITED ARGON ATOMS BY HYDROGEN AND FORMATION OF ArH^+ IONS

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A mass spectrometer with a double-chamber ion source was used to investigate the scattering of highly excited argon ions by hydrogen. The total scattering cross section was found to be $\geq 4 \times 10^{-14}$ cm^2/molecule. It was shown that ArH^+ was formed in the collisions of highly excited argon atoms with hydrogen molecules.

ARGON atoms in highly excited long-lived states have been detected by several workers.[1,2] The purpose of the present communication is to describe the main results obtained in an investigation of the collision processes of highly excited argon atoms with hydrogen molecules. The data obtained indicate that ArH^+ ions are formed not only in ion—molecule reactions,[3] but also in collisions with neutral particles, for which the interaction probabilities are considerable.

The investigation was carried out using a mass spectrometer with a double-chamber source of ions.[2] In the first chamber, the gas was excited and ionized by electron impact. Ions and electrons were held in this chamber by magnetic and electric fields. Only neutral particles, some of them excited, penetrated into the second chamber of the ion source. As a consequence of the reactions of excited particles with unexcited ones, ions were formed in the second chamber, and these were accelerated and analyzed with a magnetic analyzer.

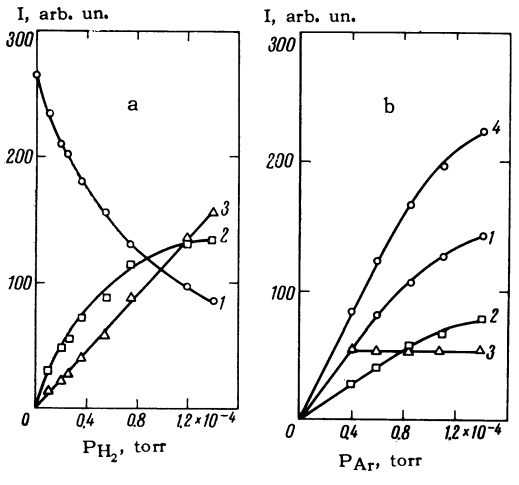

FIG. 1. Influence of the hydrogen (a) and argon (b) pressures on the intensities I of the following ions: 1) Ar^+; 2) 2) ArH^+; 4) sum of the Ar^+ and ArH^+ ion currents. Curve 3 represents the relative change in the intensities $I(ArH^+)/I(Ar^+)$. The pressure was measured with LM-2 gauge near the ion source.

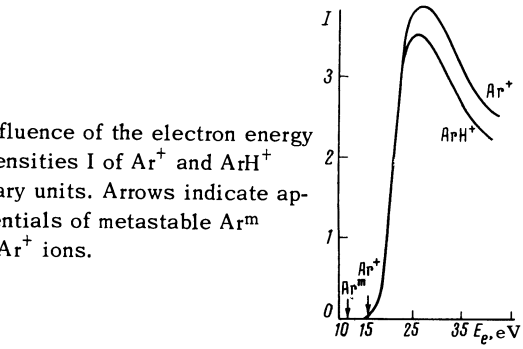

FIG. 2. Influence of the electron energy E_e on the intensities I of Ar^+ and ArH^+ ions in arbitrary units. Arrows indicate appearance potentials of metastable Ar^m atoms and of Ar^+ ions.

H^+, H_2^+, H_3^+, Ar^+, and ArH^+ ions were detected in a mixture of argon and hydrogen. To determine the mechanism of reactions leading to the formation of these ions, we investigated the dependences of the ion intensities on the hydrogen and argon pressures, as well as on the electron energy and current. Some of the dependences are given in Figs. 1 and 2.

The dependences of Fig. 1 were obtained under the following conditions. The electron energy was ≈ 27 eV, which corresponded approximately to the maximum of the yield of ArH^+ ions, as shown in Fig. 2; the ion energy was 2.8 keV; and the electron current was 0.6 mA. The argon pressure was $\approx 1.4 \times 10^{-4}$ torr (Fig. 1a), and the hydrogen pressure was 6×10^{-5} torr (Fig. 1b). The addition of hydrogen to argon (Fig. 1a) reduced the intensity of the Ar^+ ions and increased the intensity of the ArH^+ ions. The addition of argon to hydrogen (Fig. 1b) increased the intensities of the Ar^+ and the ArH^+ ions. We can see that the pressure dependences of

the relative intensities $I(ArH^+)/I(Ar^+)$ were in the form of straight lines, which was characteristic of secondary processes because $I(ArH^+) = kp_{Ar}p_{H_2}$, where k is a constant, and p_{Ar} and p_{H_2} are the pressures of argon and hydrogen.

From the data given in Fig. 2, it is evident that the ArH^+ ions appeared at an electron energy close to the energy corresponding to the appearance of the Ar^+ ions. As shown in[1,2], highly excited argon atoms Ar* appear near this energy.

In the collision of slow highly excited argon atoms with hydrogen molecules, the following main ionization processes could take place:

$$Ar^* + H_2 \rightarrow ArH^+ + H + e, \quad (1)$$

$$Ar^* + H_2 \rightarrow Ar + H_2^+ + e. \quad (2)$$

The first process was more likely; this is because the intensity of the ArH^+ ions was approximately 10 times stronger than that of the H_2^+ ions. Both reactions were exothermal. The energy evolved was transformed into the vibrational excitation of molecular ions and into the kinetic energy of electrons.

Apart from these processes, giving rise to ions, there could also be processes involving excitation energy transfer from an argon atom to a hydrogen molecule. This would produce excited molecules and fast hydrogen atoms. All these processes could be represented as the decay of an excited ArH_2 complex along various channels. Highly excited argon atoms could be deactivated by collision with a molecule, resulting in the emission of a photon, and they could also suffer elastic scattering. The probabilities of all these processes were unknown. However, we could estimate the total cross section for the loss of highly excited argon atoms from a beam in the case of scattering by hydrogen.

If the scattering took place while the argon atoms were traveling from the first chamber to the second, the intensity I(Ar*) of a beam of highly excited atoms reaching the second chamber was given by Eq. (3) (see below). In this case, it was assumed, for the sake of simplicity, that there was no loss of highly excited argon atoms due to spontaneous radiation. Thus

$$I(Ar^*) = I_0(Ar^*) \exp\{-\sigma_u n_u l_u\}, \quad (3)$$

where $I_0(Ar^*)$ is the intensity of the Ar* beam before the admission of hydrogen; σ_u is the total scattering cross-section for collisions of highly excited Ar* atoms with hydrogen molecules, whose density is n_u; l_u is the path in which the scattering in the ion source takes place. Some of the highly excited Ar* atoms were ionized on a metal grid near the second chamber and they become Ar^+ ions, i.e.,

$$I'(Ar^+) = \alpha I(Ar^*), \quad (4)$$

where α is a constant.

When hydrogen was admitted to the ion source, there was a corresponding increase in pressure in the mass-spectrometer analyzer. Consequently, the measured collector current $I(Ar^+)$ was given by the formula

$$I(Ar^+) = I'(Ar^+) \exp\{-\sigma_a n_a l_a\}, \quad (5)$$

where σ_a is the total scattering cross-section for collisions of Ar^+ ions with hydrogen molecules, whose density in the region of the mass spectrometer analyzer is n_a; l_a is the effective path in which scattering takes place.

Thus, the recorded current of Ar^+ ions should vary with hydrogen pressure as follows

$$I(Ar^+) = I_0(Ar^+) \exp\{-\sigma_a n_a l_a - \sigma_u n_u l_u\}, \quad (6)$$

where $I_0(Ar^+) = \alpha I_0(Ar^*)$. Hence, we found the total scattering cross section for collisions of highly excited argon atoms with hydrogen molecules:

$$\sigma_u = \frac{\ln[I_0(Ar^+)/I(Ar^+)]}{l_u n_u} - \frac{n_a l_a}{n_u l_u}\sigma_a. \quad (7)$$

This treatment was in accordance with the data in Fig. 1. Assuming the second term in Eq. (7) to be small compared with the first, i.e., neglecting the scattering of Ar^+ ions by H_2 molecules in the analyzer, we could estimate from Fig. 1 the upper limit of the value of the scattering cross section σ_u. Such an estimate gave the value $\sigma_{max} \sim 1.3 \times 10^{-13}$ cm^2/molecule.

We had no exact data on the value of σ_a. An analysis of the published data,[4,5] showed that, evidently, $\sigma_a \lesssim 2 \times 10^{-15}$ cm^2/molecule for Ar^+ ions of 2.8 keV energy. In this case, $\sigma_u \gtrsim 0.4 \times 10^{-13}$ cm^2.

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