USE OF EXCITED ATOMS TO STUDY THE Ar COMPONENT IN A MOLECULAR BEAM OBTAINED FROM AN Ar—He MIXTURE

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The Ar component in a molecular beam obtained in the discharge of a binary mixture of Ar and He from a nozzle source is investigated with a time-of-flight method. The excited Ar^* atoms formed by electron impact are recorded. It is shown that for an appropriate choice of recording-system parameters the contribution to the total signal from the mixture comes mainly from the Ar^* atoms. The method described is used to study the dependence of the intensity, velocity, and enrichment factor of Ar on the Ar concentration in the starting mixture.

One way to obtain intense molecular beams with velocities greater than thermal is to use a binary mixture of heavy and light gases discharging into a vacuum through a nozzle or orifice [1-3]. In order to estimate the parameters of the beams obtained in this way it is necessary to study each component of the mixture separately. It is noted in [4] that it is possible in principle to study each component separately by recording excited atoms with relatively long lifetimes ($\sim 10^{-4}$ sec) if the excited levels for the different components differ appreciably in energy. However, no data on the practical realization of this idea appear in the literature.

We separate excited Ar^* atoms from an Ar^*—He^* mixture by choosing the parameters of the excitation and recording systems in such a way as to make them sensitive to Ar^* and insensitive to He^*. This choice is based on the following: the difference in the transverse momenta transferred by electrons in inelastic interactions with Ar and He atoms; the difference in the excitation functions of Ar^* and He^* by electron impact; the difference in lifetime of the excited atoms. The contribution of each of these factors to the total ratio of the Ar^* to the He^* signals was determined experimentally.

The experiments were performed in a vacuum chamber with a gasdynamic molecular beam source. Figure 1 shows a block diagram of the experimental arrangement. A detailed description of the vacuum system, the recording system, and the signal storage is given in [5].

The molecular beam is modulated by the mechanical chopper 1 and excited by the Pierce gun 2 with the electrons moving at right angles to the axis of the molecular beam. The width X of the electron beam was 20 mm and the width Y of the entrance aperture in the direction of motion of the electrons was 5 mm. A type VÉU-1A secondary electron multiplier 3 was used to detect excited atoms. The VEU was protected against ions by attracting them to a separate collector, and against electrons by applying a negative potential of 2.6 kV to the VÉU input grid. The VÉU was mounted on a mechanism which could be displaced by an angle α in the XY plane. The angular resolution of the detector $\approx 1.5°$ was ensured by an 8-mm-diameter entrance aperture in front of the VÉU at a distance of 280 mm from the excitation region. The parameters of the molecular beam in the ground and excited states states were determined by a time-of-flight method.

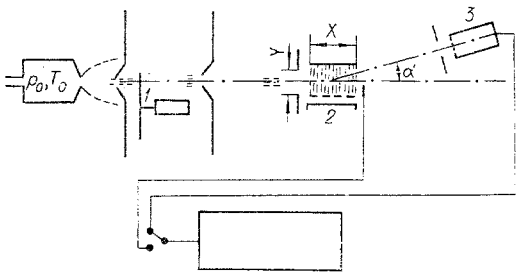

Fig. 1

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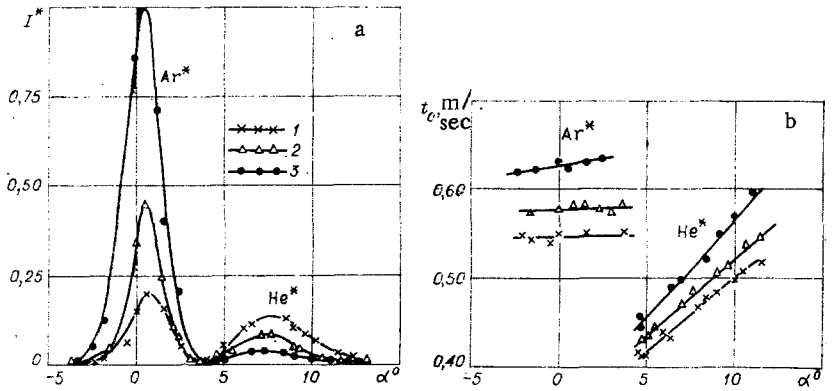

Fig. 2

To determine the contribution to the total signal from the VÉU from photoemission LiF filter was placed between the electron gun and the VÉU. This filter transmits radiation with a minimum wavelength of 1000 Å and protects the input to the VÉU against excited atoms. Measurements showed that the signal from protons was hardly noticeable against the noise background.

Since the amplification factor of the VÉU decreased somewhat after tens of hours of operation, the amplification factor of the whole system was checked periodically for fixed values of the forechamber parameters, and appropriate corrections of the results were made if the amplification changed.

The factors on which the choice of recording system parameters was based to obtain a maximum ratio of the Ar^* and He^* signals are given below.

When the electron and molecular beam intersect, the direction of the maximum of the beam of excited particles is displaced from the axis of the unexcited beam as a result of the transverse momentum (recoil momentum) transferred to the particles during excitation. Such displacements are difficult to calculate, since the spatial distributions of electrons scattered after the interaction is unknown. According to results obtained for H^* [6] and estimates made for Ar^* and He^* [7] the angular displacements for light gases can be considerable (5-15°), but for heavy gases they are much smaller ($\sim 1°$). We have measured the angular distributions of intensities I^* and the average times of flight t_0 for pure Ar^* and He^* and mixtures of them. The amplitude of the time-of-flight signal was used as a measure of the intensity. The average time of flight was determined from the position of the maximum of the signal.

The following results were obtained for pure gases: the maximum of the angular distribution of the intensity of excited atoms is displaced relative to the axis of the unexcited beam by an angle α_M varying from 0.5 to 1° for Ar^* and 8-12° for He^* for temperatures T_0 in the forechamber from 20 to 500°C. For He^* α_M obeys the law $\alpha_M V = \text{const}$, where V is the velocity of the beam. For Ar^* α_M is of the order of the error in measuring the angle and therefore it is difficult to establish any law. The intensity of He^* in the direction of the maximum of Ar^* is no more than 0.05 of its maximum value at all velocities.

The study of the angular distributions of He^* for various electron energies E showed that in a range of E not much above the excitation energy of He^* ($E^* = 23.1$ eV) the values of α_M correspond to angles obtained from Newtonian vector diagrams [6] under the assumption that the electrons are uniformly scattered in all directions. For $E > 50$ eV the position of the maximum coincides with the value obtained for the model of small angle scattering of electrons.

The results obtained for pure gases indicate that in exciting a beam formed of an Ar—He mixture a spatial separation of Ar^* and He^* atoms will occur. Figure 2 shows the angular distributions of I^* and T_0 for Ar—He mixtures with various concentrations γ of argon: 1) $\gamma = 0.003$; 2) $\gamma = 0.006$; 3) $\gamma = 0.01$ for $t_0 = 500$°C and $p_0 = 100$ mm Hg. In this case the Ar^* and He^* components are identified by comparing the positions of the intensity maxima and values of the time of flight for pure gases and mixtures. The two components are clearly

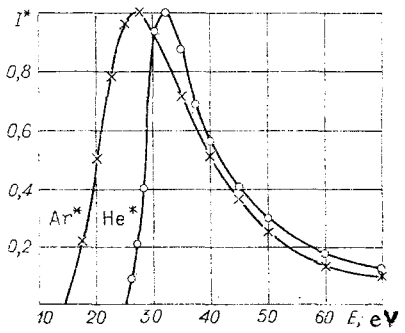

Fig. 3

separated in space; the angular distributions of t_0 show the selective effect of recoil momentum on particles with different velocities. Particles of the same kind having smaller velocities are deviated by larger angles. The effect of recoil momentum on the distribution of velocities is significantly less for Ar* than for He*, and in estimates not requiring high accuracy this effect can be neglected.

Thus, the spatial distribution of excited atoms of Ar* and He* can be used to study the Ar component in a molecular beam obtained from an Ar—He mixture.

To determine optimum conditions of excitation of each gas the dependence of I^* on E (the excitation function) was studied for a constant emission current I_e. The I*(E) curves were taken at those positions of the VEU which corresponded to the maxima of the angular distributions of intensity for E = 30 eV and T_0 = 500°C; i.e., for He* $\alpha_M = 8°$ and for Ar* $\alpha_M = 0.5°$. The excitation function curves are shown in Fig. 3. For convenience of comparison each curve is normalized to its maximum. The maximum values of the He* and Ar* signals for the same forechamber parameters are in the ratio of 1:3. This may be due to the difference in lifetime of excited atoms, to the values of the secondary emission coefficients of the VEU, or to the cross sections for excitation by electron impact. It is clear from Fig. 3 that values of E for the excitation of the mixture can be chosen so that the signal from Ar* will be appreciably larger than that from He*.

Thus the signal from the Ar* in the Ar*—He* mixture can also be separated by using the difference in the Ar* and He* excitation functions if the operating conditions prevent the use of the spatial separation of the components, or the detector does not have a sufficient angular resolution.

The finite lifetime of excited atoms leads to the deformation of their time-of-flight curve in comparison with the time-of-flight distribution of unexcited atoms. To reconstruct the time-of-flight distribution of unexcited atoms it is necessary to know the number of excited levels making the main contribution to the recorded signal and their mean lifetimes.

If the main contribution to the signal comes from excited atoms with the same value of τ the dependence of the signal amplitude I* on the time of flight between the electron gun and the VEU can be written in the form

$$I^* = aI_0^* \exp\{-t_1/\tau\}, \qquad (1)$$

where I_0^* is the intensity of excited atoms at the boundary of the excitation region closest to the VEU, and a is a constant. By varying $t_1 = l_0/V$ by changing the beam velocity V or the distance l_0 between the electron gun and the VEU it can be discovered whether Eq. (1) is satisfied, and τ can be found.

Since the area of the entrance aperture of the VEU is comparable with the cross-sectional area of the beam, a change in l_0 leads to a further dependence $I^*(l_0)$ because of the divergence of the beam, and this is difficult to take into account. Therefore t_1 was varied by changing the beam velocity V which is a function of the temperature T_0 of the gas in the forechamber. However, the intensity I_0 of the beam of atoms in the ground state also depends on T_0, and, consequently, I_0^* does also. This dependence was eliminated by investigating $K(t_1) = I^*/I^+$ instead of $I^*(t_1)$, where I^+ is the signal from the ion collector when the electron gun operates in the regime of an ionization detector, which was achieved by increasing the energy of the electron beam to 150 eV. For excitation and ionization by an electron beam we can write $I_0^* = bI_0$ and $I^* = cI_0$, where b and c are excitation and ionization coefficients which depend on V or T_0 in the same way. Consequently, the relation $K(t_1)$ gives the required function to within a constant factor.

It turned out that ln K is a linear function of t_1 within the limits of experimental error for both Ar* and He*. Hence it follows that the excited atoms arriving at the VEU

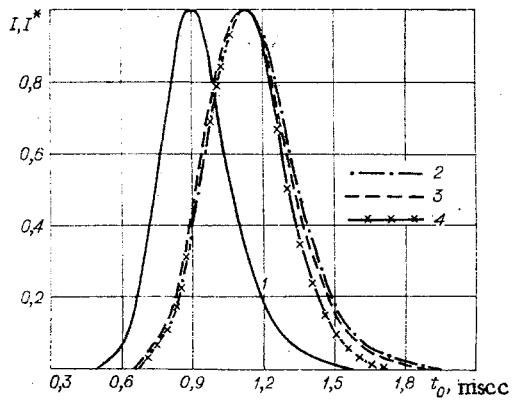

Fig. 4

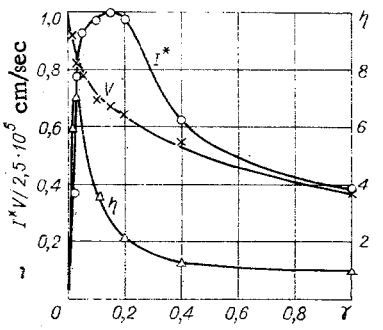

Fig. 5

are mostly atoms with the same lifetime τ which can be determined from the relation $\ln K(t_1)$. The experimental values of τ for Ar* and He* are $\tau_{Ar} = (1.8 \pm 0.3) \cdot 10^{-4}$ sec and $\tau_{He} = (1 \pm 0.2) \cdot 10^{-4}$ sec. For values of E from 25 to 35 eV the value of τ_{Ar} is independent of E.

Electron transitions from excited levels were identified by the measured lifetimes. According to [8-9] the transition $5p\ [1^1/_2] \rightarrow 4s'\ [^1/_2]^\circ$ for Ar* has a lifetime of $1.9 \cdot 10^{-4}$ sec, which is close to the experimental value. This corresponds to a transition from a state with energy $E_2 = 14.53$ eV to a state with energy $E_1 = 11.83$ eV. There is a transition from the $4s'\ [^1/_2]^\circ$ level to the ground state $sp^6\ ^1S_0$ during a time of $1.4 \cdot 10^{-9}$ sec [10]. Thus, the main contribution to the VEU signal for Ar* comes from the excited level with the energy 14.53 eV from which there occurs a cascade of transitions to the ground state. This is confirmed also by the value of the threshold exciting energy ≈ 14.5 eV which can be obtained at the point of intersection of the I*(E) curve with the axis of abscissas.

A number of other transitions with lifetimes $\sim 10^{-4}$ sec can be found in the tables of [8-9] but they have small relative intensities. Taking account of transitions with lifetimes closest to that of the main transition with appropriate weights depending on intensity gives the calculated value $\tau_{Ar} = 1.81 \cdot 10^{-4}$ sec. The tabulated value [11] of the He lifetime closest to the experimental value is $\tau_{He} = 0.8 \cdot 10^{-4}$ sec and corresponds to the transition $3^3D \rightarrow 3^3P_0$.

The validity of the determination of τ for Ar* and the effect of the error in determining τ on the parameters of the time-of-flight signal were estimated. The experimental time-of-flight curve of the Ar^+ ion current (Fig. 4, curve 1) was used to calculate curves of the Ar* signals from the VEU, taking account of the value obtained for τ. Two values of τ were used in the calculations: the average value $\tau = 1.8 \cdot 10^{-4}$ sec (curve 2) and the minimum value $\tau = 1.5 \times 10^{-4}$ sec (curve 3) lying within the limits of experimental error. The calculated curves were compared with the experimental time-of-flight curve 4 obtained in recording Ar*. The curves calculated for $\tau = 1.8 \times 10^{-4}$ sec and $\tau = 1.5 \cdot 10^{-4}$ sec differ insignificantly from one another. The curve for the measured Ar* signal differs slightly from the calculated curves for small t (large velocities). For large t the experimental curve shows a deficiency of slow particles, an effect which increases with increasing t. This can be accounted for by the effect of recoil momentum produced by the preferential deviation of slow atoms from the axis of the beam. The deficiency of slow atoms in the measured Ar* signal will have little effect in determining the total flux (the areas under the experimental curves is less than the area under the calculated curve by 5%). The positions of the maxima of the experimental and calculated curves practically coincide; a significant difference is observed only in the half-widths where the difference between the measured and calculated curves is 10%.

Thus with the method used to determine τ the time-of-flight distribution of atoms in the ground state can be determined with sufficient accuracy from the shape of the time-of-flight curve for the excited atoms.

We note that the difference in lifetime of Ar* and He* can also be used to separate the Ar* signal from that of the mixture. If we assume [3] that the velocities of the two components of the mixture are the same and equal to V, the ratio of the Ar* intensities is

$$q = \frac{I^*_{Ar}}{I^*_{He}} = B \exp\left\{-\frac{l_0}{V}\left(\frac{1}{\tau_{Ar}} - \frac{1}{\tau_{He}}\right)\right\},$$

where B is a constant. By increasing the distance l_0 between the electron gun and the VÉU for fixed V, τ_{Ar}, and τ_{He} a large enough value of q can be obtained.

The ratio of the Ar* and He* signals in the mixture was estimated, taking account of the effect of recoil, the excitation functions of Ar* and He*, and the lifetime of excited atoms with the detector in the position corresponding to the maximum of the Ar* angular distribution. For a 10% concentration of Ar in the forechamber this estimate gave a minimum ratio of $\sim$ 15, but the measured value turned out to be much higher.

The recording method described was used to determine the composition of the Ar–He mixture which would ensure a relatively high intensity of the molecular beam of Ar for velocities of Ar atoms close to the velocities of the He atoms. To do this the intensities and average velocities of beams of Ar, He, and Ar–He mixtures with various concentrations of Ar were measured. In all the measurements of this series the axis of the VÉU coincided with the axis of the unexcited beam, since the angle of deflection of the maximum of the Ar* beam ($\alpha_M \simeq 0.5°$) was smaller than the angular resolution of the VÉU. The velocity of a beam of pure He was determined from the time-of-flight signal of the ion current. The velocity and intensity of Ar in the mixture were determined from the Ar* signal.

The signals from the mixture with various concentrations of Ar were from 30 to 100 times larger than the signal from pure He*. Since the density and velocity of He are lower in the Ar–He beam than in the beam of pure He for the same values of p_0, the relative contribution of He* to the total signal from the mixture will be still less than the ratio of the measured signals for pure gases. On the basis of this the contribution of He* to the total signal from excited atoms of the mixture can be neglected and it can be assumed that the signal from Ar*–He* is due entirely to Ar* atoms.

Figure 5 shows the dependence of the intensity I* (in relative units) and the velocity V of the Ar* beam on the concentration γ of Ar in the starting mixture for a pressure p_0 = 100 mm Hg and a temperature T_0 = 500°C in the forechamber. For values of γ = 0.03–0.1 the velocity of Ar is close to the maximum velocity of He. The recorded signal in this case is significantly higher than for pure Ar. This cannot be accounted for solely by the decrease in the time of flight but indicates an increased concentration of argon in the region close to the axis of the beam. To estimate the increase of the Ar intensity in the mixture in comparison with the calculated values we introduce the enrichment factor $\eta = I_m/I_c$, where I_m is the measured intensity of Ar in the mixture and I_c is the calculated intensity taking account only of the increase in velocity of Ar due to acceleration by helium.

From the dependence of η on γ shown in Fig. 5 one can judge the increase in the relative concentration of Ar on the axis of the beam in comparison with the concentration in the forechamber.

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