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Abstract

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SEPARATION OF ISOTOPES DURING THE PASSAGE OF AN ATOMIC BEAM THROUGH AN IONIZATION SPACE

1. Simple theoretical considerations indicate the possibility of separating isotopes during the passage of an atomic beam, consisting of a mixture of isotopes, through a space in which it is subjected to bombardment by electrons. Suppose that the atomic beam consists of particles with masses m_1 and m_2, with $m_1 < m_2$, and emerges from a region having temperature T; then the mean kinetic energy of the particles will be ${}^3/_2\, kT$, and

$$v_1/v_2 = \sqrt{m_2/m_1},$$

where v_1 and v_2 are the root-mean-square velocities of the corresponding isotopes.

Let us now assume that such a beam is penetrated by electrons with energy sufficient for its ionization, and that the ions formed are continuously removed from the beam. Then the attenuation of the atomic beam will occur according to the following law:

$$J(x + \Delta x) = J(x) - \frac{J(x)}{v}\, j\sigma \Delta x, \tag{1}$$

where $J(x)$ and $J(x+\Delta x)$ are the atom flux densities, respectively, at the point x and $x+\Delta x$; v is the mean velocity of the atoms in the beam, and consequently $J(x)/v = \rho$ is the number of neutral atoms per unit volume; j is the electron current density; σ is the ionization cross section of the atoms for electrons of a definite energy.

Integrating expression (1), we obtain:

$$J(x) = J(0) \exp\left[-\frac{j\sigma}{v} x\right], \tag{2}$$

where $J(0)$ is the density of the atomic beam before passing through the ionization region.

Let us denote by J^1 and J^2 the densities of the atomic beams of particles with masses m_1 and m_2. Thus, before entering the ionization space, the ratio of the concentrations of particles of different mass is equal to $J^1(0)/J^2(0)$.

After the beam has passed through a distance x in the ionization space, the ratio of the concentrations will be expressed as follows:

$$\frac{J^1(x)}{J^2(x)} = \frac{J^1(0)}{J^2(0)} \exp\left[-j\sigma\left(\frac{1}{v_1} - \frac{1}{v_2}\right)x\right], \tag{3}$$

where it is assumed that the effective ionization cross sections σ_1 and σ_2 of the isotopes are approximately the same, i.e. $\sigma_1 = \sigma_2 = \sigma$. Substituting the expression v_1/v_2 into (3), we obtain

$$\frac{J^1(x)}{J^2(x)} = \frac{J^1(0)}{J^2(0)} \exp\left[-\frac{j\sigma}{v_1}\left(1 - \sqrt{\frac{m_2}{m_1}}\right)x\right], \tag{4}$$

or, expanding the expression $\sqrt{m_2/m_1}$ in a series and retaining only the first term of the expansion:

$$\frac{J^1(x)}{J^2(x)} = \frac{J^1(0)}{J^2(0)} \exp\left[\frac{i\sigma}{v_1}\frac{\Delta m}{m_1}x\right]. \tag{5}$$

From formula (2), for a beam with mass m_1, we find:

$$\frac{i\sigma}{v_1} = -\ln\frac{J^1(0)}{J^1(x)}. \tag{6}$$

Substituting (6) into (5), we obtain:

$$\frac{J^1(x)}{J^2(x)} = \frac{J^1(0)}{J^2(0)} \exp\left[\frac{\Delta m}{m_1}\ln\frac{J^1(0)}{J^1(x)}\right], \tag{7}$$

or

$$\frac{J^1(x)}{J^2(x)} = \frac{J^1(0)}{J^2(0)}\left(\frac{J^1(0)}{J^1(x)}\right)^{\Delta m/m_1}. \tag{8}$$

It follows from expression (8) that, as a result of attenuation of the beam, its composition should change in the direction of enrichment with light particles.

Fig. 1. 1 –ampoule with mercury; 2 –valve; 3 –LM-2 tubes

Figure 1: Fig. 1. 1 –ampoule with mercury; 2 –valve; 3 –LM-2 tubes

2. To detect this effect it was necessary to use a system which would make it possible, on the one hand, to ionize the passing atomic beam and, on the other, to remove the ions formed and bind them. These requirements are satisfied by the LM-2 triode manometer tube.

When an atomic beam passes through the LM-2, it is ionized by electrons emitted by the cathode and accelerated by the grid potential. In this case, the ionization region is the space inside the grid and outside it over the distance in which the electrons that have passed through the grid are slowed down to an energy equal to the ionization potential of the substance under study.

Fig. 1. 1 –ampoule with mercury; 2 –valve; 3 –LM-2 tubes

The distribution of potentials in the LM-2 ($V_{\text{anode–cathode}} = -25$ V, $V_{\text{anode–grid}} = -225$ V) is such that the ions must be drawn to the anode and cathode. Ions reaching the cathode will be re-evaporated, while ions reaching the anode will remain on it, which will cause attenuation of the passing beam and a change in its composition. This can be detected by analyzing the beam on a mass spectrometer.

Figure 1 shows a system consisting of five LM-2 tubes connected in series, which we used to detect and investigate the phenomenon described. Mercury was taken as the working substance; its vapor could pass successively through the entire system of five tubes. During conditioning of the tubes the vessel with mercury was shut off, and the supply of mercury vapor to the system was stopped. During the experiment the mercury had a temperature of $\sim 20^{\circ}$, the glass of the tubes $\sim$ 50–60°, and the nickel leads $\sim$ 100–120°. The pressure in the tubes during the experiment did not exceed $\sim 2 \cdot 10^{-5}$ mm Hg. The tubes were powered by five separate vacuum meters of the VI-3 type. The mercury passing through the tubes was analyzed with an MS-2 mass spectrometer. The measurements were carried out by a single-beam method on the extreme isotopes of mercury (198 and 204), i.e., the ion currents corresponding to iso-

isotopes 198 and 204, and then their ratio was calculated. The measurements were carried out in series of 10 ratios, from which the arithmetic mean was calculated. The measurement time for one series was about 2 min.

The attenuation of the mercury-vapor flux (the experiments were carried out with a mercury-vapor flux, and not with an atomic beam) was calculated from the ratio of the isotope-198 peaks before the start of ionization and during ionization. The composition of the mercury-vapor flux was measured alternately with the lamps switched off (standard composition) and with them switched on (altered composition). It had first been verified that no operating mode of the lamp, except that in which gas ionization takes place, changes the isotopic

Fig. 2

Figure 2: Fig. 2

Fig. 3

Figure 3: Fig. 3

composition of the mercury flux.

Table 1

Measured	Calculated
0.62 ± 0.34	0.34 ± 0.14
0.62 ± 0.28	0.28 ± 0.14
0.69 ± 0.34	0.42 ± 0.14
0.62 ± 0.34	0.34 ± 0.14
0.76 ± 0.34	0.48 ± 0.14
1.58 ± 0.28	1.17 ± 0.14

3. Table 1 gives the results of measurements and calculations of the enrichment Δ of the outgoing flux. The measurements were carried out in turn on each of the five lamps and when all five lamps were switched on successively (the last row of Table 1).

$$\Delta = \frac{J^1(x)/J^2(x) - J^1(0)/J^2(0)}{J^1(0)/J^2(0)} \cdot 100\%.$$

Calculations by formula (8) of the values of Δ were performed on the basis of experimental data on the attenuation of the flux.

Comparison of the experimental and calculated data shows their good agreement within the limits of the measurement errors; however, it should be noted that all measured values are larger than the calculated ones. The deviation of the measured data from the calculated data is most noticeable when all the lamps are switched on simultaneously (the last row of Table 1). As to the possible reasons for such a discrepancy, only some suppositions can be put forward.

Fig. 2. Testing five LM-2 lamps for saturation with mercury with periodic checking of the standard composition

Fig. 3. Desorption of mercury from the anodes of LM-2 lamps

First of all, it should be taken into account that in the ionization region there exists molecular flow of mercury vapor, and not an atomic beam, and, consequently, mercury atoms can collide with the walls and electrodes of the lamp and pass many times through the ionization region. Further, the calculation did

not take into account the difference in the velocities of the beam atoms; however, estimates made of the error due to this (assuming a normal Maxwellian velocity distribution) gave a correction 10 times smaller than the deviation of the calculated data from the measured data.

Measurements were carried out with the lamp system (Fig. 1) in order to determine their saturation time. Figure 2 presents two graphs: the deviation of the beam composition from the standard as a function of time, and the change with time of the beam of isotope 198. During periodic checking of the standard composition, bursts of intensity are observed on the upper curve, indicating how much the ionization weakens the mercury beam. From the graphs it is evident that the enrichment gradually decreases, as does the attenuation, although even after 20 hours of operation the beam is enriched by an appreciable amount.

As a methodological remark, it should be noted that, during operation of the lamps, changing the beam (by changing the mercury inlet by approximately 1/3) did not lead to a change in its composition. This is seen from the burst of intensity on the upper curve and from the constancy of the composition on the lower curve during the period from 6 to 7 hours (Fig. 2). The composition of the mercury desorbed from the lamp anodes after many hours of operation was also checked. Desorption from the anodes was produced by switching the lamps on for "warming" in the absence of mercury vapor entering from the ampoule. As a result of the warming, a sharp increase in intensity was observed, while the composition of the desorbed mercury was below the standard; i.e., the composition shifted toward a higher content of the heavy isotope. Figure 3 presents two graphs: the changes in intensity and in the composition of the desorbed mercury.

It should be noted that the phenomenon described may be a source of serious systematic errors in precise mass-spectrometric measurements.

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Note: Figure translations are in progress. See original paper for figures.