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Design considerations of a two-stage alkali atom beam source with a multicollimator*

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The paper contains the design considerations of a two-stage source of alkali atoms beam suitable for studies of multiphoton processes. The construction of the source makes it possible to obtain a high intensity beam of small divergence and reduced concentration of dimers with a small consumption of mass of the material used.

1. Introduction

A method of atomic and molecular beams has found many applications in various fields of physics. It is of particular advantage in the optical studies, e.g., in Doppler-free spectroscopy and multiphoton processes. The simplest source of the atomic or molecular beam is an effusion oven with a thin circular hole. According to RAMSEY [1], the number dN of atoms effused from the orifice within a solid angle $d\omega$ at an angle ϑ measured with respect to the direction of the beam axis is

$$dN = (1/4\pi)\, d\omega n \bar{v} A \cos\vartheta \tag{1}$$

where: n – density of atoms in the oven, A – area of the orifice, $\bar{v}$ – mean velocity of an atom which at Maxwell distribution is $\bar{v} = \sqrt{8kT/\pi m}$, m – mass of an atom.

Total number of atoms leaving the source during time unit is

$$N = (1/4)\, n \bar{v} A, \tag{2}$$

so the number of atoms flowing in the desired direction $\vartheta \simeq 0$ is small when compared with the total effusion rate. This leads to disadvantageous wastage of the metal placed in the furance and gives rise to problems with external collimation of the atomic beam. One of the methods of avoiding of the losses is the recirculation of the material effused from the furnace within nonaxial part of the beam [2]–[4]. Another one consists in the application of a pipe of a length l instead of an outlet hole. Assuming effusive outflow from the channel, i.e., when the

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mean free path of an atom is

$$\lambda = (\sqrt{2}\pi\delta^2 n)^{-1} \gg l \tag{3}$$

where δ is the effective diameter of an atom, the number of atoms leaving the source can be calculated from Eq. (2) into which the geometry reduction coefficient (Clausing coefficient) has to be incorporated, so

$$N = \frac{1}{4\varkappa} n\bar{v}A. \tag{4}$$

In addition, the dependence of the angular distribution on the ratio l/a [5] has to be taken into account as well. Condition (3) seriously limits the brightness of the source.

According to this condition, in order to obtain the required geometry of the beam, the mean free path of an atom should be increased or the length, so the diameter also, of the channel should be decreased, which may be unfeasible. An increase in the peak intensity of the atom beam, with its geometry unchanged, can be realized by replacing the effusion flow with ultrasonic one, or in the case when atoms of thermal velocity are required – by applying a multicollimator made of many parallel effusive nozzles.

As it was already found by HELD et al. [6] the presence of even slight admixture of alkali dimers in the atomic beam can be a source of significant errors, while studying multiphoton phenomena. When vapour is in thermodynamic equilibrium with the surface of the melted alkali metal, partial pressure of dimers considerably increases with temperature, as shown in Fig. 1. Although the rate of dimers dissociation increases with temperature, an increase in vapour density

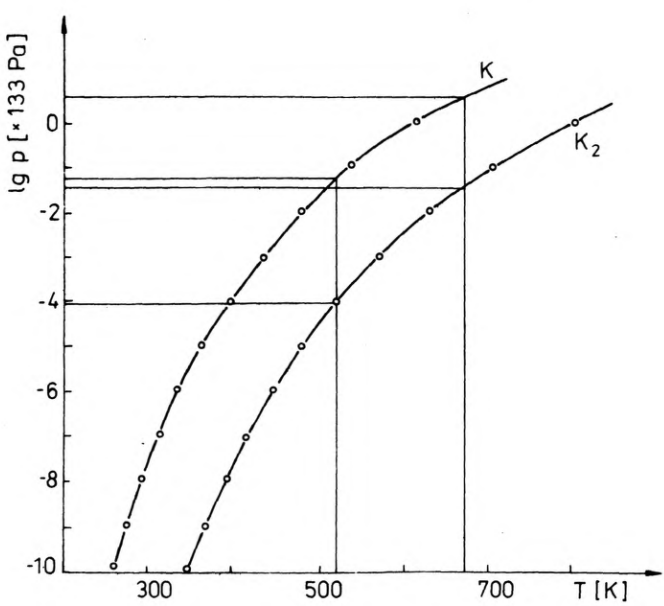

Fig. 1. Partial pressures of potassium atoms and dimers as function of temperature. Data points after [13]

under thermal equilibrium with the melted metal leads to the domination of dimers formation over their dissociation.

In order to diminish the contribution of dimers in the beam, a separated region of higher temperature should be introduced so that it should not be in thermal equilibrium with the melted metal. As it follows from the above considerations the source of alkali atom beams designed, for example, to study multiphoton processes should consist of a container of alkali metal, where a stream of vapour of a required density is produced, a superheater to assure effective dimers dissociation, a tube connecting the container with the superheater and a multicollimator nozzle which provides high intensity and required angular distribution. The dimensions of the tube are selected so that the atoms undergo many collisions with walls and only few collisions with themselves.

The sources of this kind have already been applied in the studies of multiphoton ionization (e.g., [7]), however they have not been described in detail so far.

2. Flow of atoms through long pipes

Let us consider the flow through a tube of a radius a and length $l \gg a$. One end of the tube is assumed to be in the vacuum and the other is connected with a gas container of a density n. Let us further assume that the atoms reflected from the walls of the pipe obey Knudsen's law [8].

As to the relation between the length and radius of the pipe and the average mean free path $\bar{\lambda}$ of an atom there can be distinguished in the pipe, after GIORDMAINE and WANG [9], three kinds of flow.

Mode I flow: $\bar{\lambda} \gg l$, the atoms undergo collisions with the walls of the pipe only and the influence of interatomic collisions is negligible. The parameters of the beam can then be described by the following relations:

peak intensity

$$I(0) = \frac{1}{4} n \bar{v} A \left[\frac{\text{atom}}{\text{sr s}}\right], \tag{5}$$

total flow intensity

$$N = \frac{1}{4\varkappa} n \bar{v} A = \frac{\pi}{\varkappa} I(0) \left[\frac{\text{atom}}{\text{s}}\right], \tag{6}$$

Clausing coefficient

$$\varkappa = 1 + \frac{3l}{8a}, \quad \text{for } l/a \geqslant 1.5,$$
$$\varkappa = \frac{3l}{8a}, \quad \text{for } l \gg a. \tag{7}$$

In order to obtain more intense beams the condition $\bar{\lambda} \gg l$ is often replaced by a weaker one.

Mode II flow: $\bar{\lambda} \leqslant l$ and $\lambda \gg a$ in the whole tube. Physically it means that the interatomic collisions inside the channel cannot be completely neglected but their influence does not alter the distribution of collisions of the atoms with the tube walls. In this case, Eq. (6) is still valid although the peak intensity is not proportional to the pressure in the source. The shape of the beam depends on both geometry of the pipe and flow intensity N.

Since the quantity $I(0)$ does not provide any information about directional properties of the source and the dependence $I(\vartheta)$ is complicated, we consider it purposeful to introduce the Clausing coefficient [10] in a more general sense as

$$\varkappa^* = \left.\frac{I(0)_{\text{tube}}}{I(0)_{\text{orifice}}}\right|_{N=\text{const}} \tag{8}$$

Thus, the $\varkappa^*$ coefficient determines how many times the intensity in the beam axis for the source with a pipe is greater than the intensity from the source with a diaphragm of the same diameter as the pipe under constant total flow. Simultaneously, the value of this coefficient is a convenient measure of directional properties of the source. As it follows from the theory of GIORDMAINE and WANG [9]

$$I(0) = \frac{1}{\pi} \varkappa^* N, \tag{9}$$

for any mode of molecular flow.

For the flow of II Mode

$$\varkappa^* = \frac{\sqrt{3\pi}}{8 \cdot 2^{1/4}} \frac{1}{\delta} \sqrt{\frac{\bar{v}a}{N}} \tag{10}$$

and

$$I(0) = \frac{2^{1/4}}{8} \frac{\bar{v} a^2 n^{1/2}}{\delta l^{1/2}}. \tag{11}$$

It is worth noting that the method of increase in the peak intensity $I(0)$ by increasing pressure is ineffective rather, because of proportionality to $\sqrt{n}$. For the sake of approximation, the formulae (10) and (11) can be applied up to the value $\lambda \geqslant a$.

Mode III flow: $\bar{\lambda} \ll l$. The flow of Knudsen type occurs only to some distance from the outlet, and in the remaining part of the tube the flow of Poisseuille type. For the flow of Mode III it is hard to obtain good collimation as only a part of the tube participates effectively in the collimation process.

All the above formulae were derived on neglecting the influence of low-angle collisions between the atoms (the effective cross-section of low-angle collisions can

be significantly larger than the gaseous-kinetic cross-section), of diffusion of atoms on the tube wall, and of the effect of choking of the flow by the outlet of the tube (implied by a nonzero pressure at the outlet due to the cloud of atoms). The influence of these factors was discussed by BECKER [10] and leads to the corrected value of $\varkappa$ coefficient.

In the region $\bar{\lambda} < a < 100\bar{\lambda}$ which is important for the tube connecting the container with the superheater one may apply neither Eq. (6) for the effusive flow nor equations for viscous flow. In this case we refer to the empirical equation given by ŻUK [11] obtaining

$$N' = N\left(0.1472\frac{a}{\bar{\lambda}} + Y\right) \tag{12}$$

where: $Y = \dfrac{1+0.3342X}{1+0.4126X}$, $\quad X = \dfrac{(p_1+p_2)\,a}{2\bar{\lambda}}$,

N is taken from Eq. (6) for molecular flow, and p_1 and p_2 are the pressures in Pa at both ends of the tube. We can calculate then

$$N' = \frac{1}{4\varkappa'} n\bar{v}A, \tag{13}$$

$$\varkappa' = \varkappa\left(0.1472\frac{a}{\bar{\lambda}} + Y\right)^{-1}. \tag{14}$$

3. Source design

In Figure 2 a scheme of our two-stage source of atomic beam is presented. In order to calculate its parameters we will apply the procedure similar to the one employed by LAMBROPOULOS and MOODY [2] for a source with a diaphragm. The notation used will be as follows:

p_1, p_1 – pressures in the oven and superheater, respectively,
T_1, T_2 – absolute temperatures,
n_1, n_2 – atom densities,
$A_t = \pi a_t^2$ – area of cross-section of the connecting tube,
$A_0 = \pi a_0^2$ – area of cross-section of a single channel in the multicollimator,
$\varkappa_1$ – geometrical Clausing coefficient of the connecting tube,
$\varkappa_0$ – geometrical Clausing coefficient of the one channel in the multicollimator,
$\varkappa_1'$ – Clausing coefficient from Eq. (14),
$\varkappa_0^*$ – generalized Clausing coefficient defined by Eq. (8),
M – number of channels in the multicollimator,
$\bar{v}_1$, $\bar{v}_2$ – average velocities of the atoms,
λ_1, λ_2 – mean free paths,
$N\uparrow$ – intensity of the flow up the connecting tube,

$N\downarrow$ – intensity of the flow down the connecting tube,
N_{eff} – intensity of the outflow from the multicollimator,
m – mass of the atom.

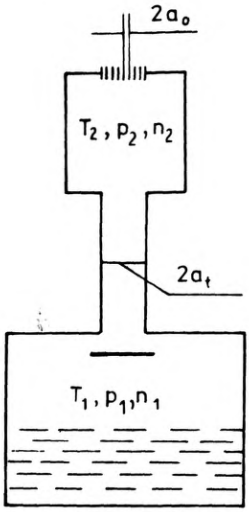

Fig. 2. Schematic outline of the two-stage oven with a multicollimator nozzle

Under the assumption that the effusion from multicollimator satisfies the conditions for the flow of I or II Mode, and the flow through the tube obeys the ŻUK criterion, we may write the following:

$$N\uparrow = \frac{1}{4}\frac{1}{\varkappa_1'} n_1 \bar{v}_1 A_t,$$

$$N\downarrow = \frac{1}{4}\frac{1}{\varkappa_1'} n_2 \bar{v}_2 A_t, \tag{15}$$

$$N_{\text{eff}} = \frac{1}{4}\frac{1}{\varkappa_0} M n_2 v_2 A_0.$$

Since $N\uparrow = N\downarrow + N_{\text{eff}}$, we obtain

$$n_2 = n_1 \frac{\bar{v}_1}{\bar{v}_2} \frac{\varkappa_1'}{1 + M\dfrac{\varkappa_1'}{\varkappa_0}\dfrac{A_0}{A_t}} = n_1 \sqrt{\frac{\overline{T_1}}{T_2}}\, g \tag{16}$$

where

$$g = \left[1 + M\frac{\varkappa_1'}{\varkappa_0}\frac{A_0}{A_t}\right]^{-1}. \tag{17}$$

Thus

$$N_{\text{eff}} = \frac{1}{4}\frac{1}{\varkappa_0}\bar{v}_1 n_1 A_0 M g = \frac{1}{\sqrt{2\pi}} p_1 \frac{1}{\sqrt{k T_1 n_1}} \frac{1}{\varkappa_0} M A_0 g. \tag{18}$$

It is worth to note that N_{eff} does not depend explicitely on the superheater temperature. In general, however, g depends on T_1. It is easy to find that

$$p_2 = p_1 \sqrt{\frac{T_2}{T_1}} g \tag{19}$$

and

$$\lambda_2 = \frac{k\sqrt{T_1 T_2}}{\sqrt{2}\pi\delta^2 p_1 g}. \tag{20}$$

Having Eq. (18) substituted to Eq. (9) one obtains for the peak intensity

$$I(0) = \frac{1}{\pi} \frac{1}{\sqrt{2\pi m k T_1}} M g A_0 \frac{\varkappa_0^*}{\varkappa_0} \tag{21}$$

where $\varkappa_0^*$ is given by Eq. (10).

Equation (21) is valid when we consider the value of peak intensity at a very long distance from the outlet of the multicollimator as compared with the diameter of the whole multicollimator.

The pressure of the saturated vapour of alkali metals can be determined from

$$\lg p_1 \ [\text{Pa}] = -\frac{A}{T} + B \lg T + CT + D \tag{22}$$

where values of the coefficients can be found from [12]. The set of Eqs. (16)–(21) allows the determination of main parameters of the source. Their application is limited by the conditions

$$\lambda_1 > 0.01 a_t \quad \text{and} \quad \lambda_2 \geqslant a_0.$$

4. Dissociation of alkali dimers

The process of dissociation of alkali dimers requires some detail considerations. The rate of molecular dissociation, which for diatomic molecules runs along the scheme $AB + C \xrightarrow{D} A + B + C$, can be determined from the equation

$$k_d = \frac{q_{ABC}}{q_{AB} q_C} \frac{kT}{2\pi\hbar} e^{-D/kT} \tag{23}$$

where q_{ABC}, q_{AB}, q_C are the statistical weights of the transient complex ABC and of its components AB and C, respectively, whereas D is the energy of dissociation of molecule. Both dissociation and association processes are competing, so also $A + B + C \rightarrow AB + C$.

The concentration of the dissociated atoms depends on the relative rate of both processes at a given temperature. The ratio of these two rates is referred to as an

equilibrium constant and is equal to

$$K_e = \frac{k_d}{k_a} = \frac{q_A + q_B}{q_{AB}} e^{-p/kT}. \tag{24}$$

In the case of thermal dissociation of the molecule consisting of two identical atoms the equilibrium constant depends only on temperature and can be expressed as

$$K_e(T) = \frac{(p_{atom})^2}{p_{dimer}}. \tag{25}$$

With regard to this, the ratio of partial pressures of dimers for different temperatures of the superheater is

$$\frac{p_d(T_2)}{p_d(T_1)} = \frac{p_a^2(T_2)}{p_a^2(T_1)} \frac{K_e(T_1)}{K_e(T_2)} = \frac{T_2}{T_1} \frac{K_e(T_1)}{K_e(T_2)} \tag{26}$$

where the values $p_a(T)$ are substituted by the pressures in the superheater calculated from Eq. (19).

The equilibrium constant can be determined on the basis of the known partial pressures in thermodynamic equilibrium. For example, taking from Fig. 1 the values of partial pressures for $T_1 = 523$ K and $T_2 = 673$ K, we find $K_e(T_1) = 4.2 \times 10^3$ Pa and $K_e(T_2) = 265.8 \times 10^3$ Pa. Thus

$$p_d(T_2)/p_d(T_1) = 0.02,$$

which points to the fact that the superheater reduces effectively the number of dimers.

5. Construction of the source

According to the scheme in Fig. 2 the source was built with the following parameters: $l_t = 40$ mm, $a_t = 1.5$ mm, $l_0 = 6$ mm, $a_0 = 0.2$ mm, $M = 80$. It was used in the studies of multiphoton ionization of atomic potassium.

All the elements coming in contact with hot metal are made of stainless steel, and the seals are made of copper. The multicollimator consists of the segments of hypodermic needles. The furnace was heated with heaters made of Kanthal wire (of 0.3 mm in diameter) wound on a ceramic tube and put inside another tube. The numbers of such heaters in the oven amounted to six, and in the copper block surrounding the superheater nine heaters were placed. Temperature was measured by thermocouples placed in the holes in the oven and the superheater.

Total power required to maintain the temperatures $T_1 = 523$ K and $T_2 = 673$ K is of about 100 W, and for $T_1 = 463$ K and $T_2 = 610$ K is reduced to 35 W. At the above temperatures the pressure in the container is $p_1 = 7.16$ Pa and the atom density is $n_1 = 9.9 \times 10^{20}$ m^{-3}. The mean free path in the container is $\lambda_1 = 1.04$ mm. Under these conditions we can avoid tedious iterative procedure

to find $\bar{\lambda}$ and $\bar{p}$ in the connecting tube, and take for the total outflow $\varkappa_1' = \varkappa_1 = 11$, with the accuracy to few percents. In this case $\bar{v}_1 = 552$ m/s, $g = 0.44$, $n_2 = 3.85 \times 10^{20}$ m^{-3}, $p_2 = 3.57$ Pa, $\lambda_2 = 2.67$ mm.

In other words, we are dealing here with the Mode II outflow from the multicollimator. Thus, we have $\bar{v}_2 = 604$ m/s, $N_{\text{eff}} = 4.76 \times 10^{16}$ atoms/s, $\varkappa_0 = 9.81$, $I(0) = 1.48 \times 10^{17}$ atoms/sr s, and at the distance of 10 cm from the multicollimator outlet the atoms density is 2.5×10^{10} atoms/cm^3. An essential parameter, for practical reasons, is the time of emptying the container of potassium. Under the conditions described above it reaches to about 90 h/g. Most of the measurements have been carried out under the following parameters:

$T_1 = 468$ K, $T_2 = 608$ K, $\lambda_2 = 24$ mm, $\bar{v}_2 = 573$ m/s,

$N_{\text{eff}} = 5 \times 10^{15}$ atoms/s,

$I(0) = 1.9 \times 10^{19}$ atoms/sr s $= 3.4 \times 10^9$ atoms/cm^3,

$Q = 860$ h/g.

Observations of dimer concentration were carried out in the set-up devoted for observation of three-photon ionization of atomic potassium [14]. Ions were generated by a strong laser field and observed with the use of time-of-flight spectrometer. In the case of ionization of atoms the following process takes place:

$$K \xrightarrow{4.85\ \text{eV}} K^+ + e.$$

The decomposition of dimers can occur in a few different ways:

$$K_2 \begin{cases} \xrightarrow{4.35\ \text{eV}} K^+ + K^- \\ \xrightarrow{4.06\ \text{eV}} K_2^+ + e \xrightarrow{0.75\ \text{eV}} K + K^+ + e \\ \xrightarrow{4.85\ \text{eV}} K^+ + K + e \end{cases}$$

only one of them giving molecular ion signals, which can be distinguished from ionization of atoms. Nevertheless, these ionic signals could be used for the evaluation of superheater effectivity. In the case when the superheater was operat-

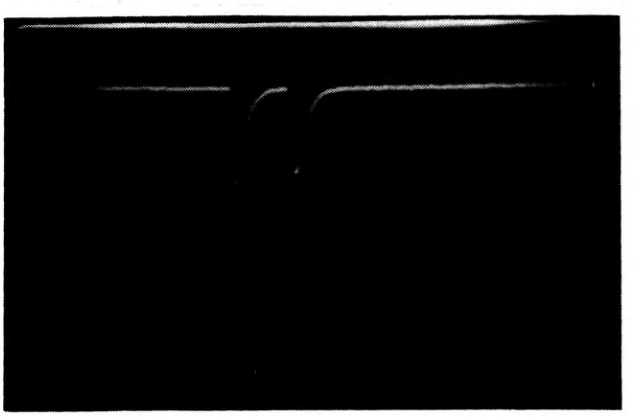

Fig. 3. Time-of-flight spectrometer signals showing atom (first peak) and dimer (second peak) ions

ing in the temperature equal to that of the furnace, additional signals due to dimers ionization were observed (Fig. 3). With increasing temperature of the superheater this additional signal was decreasing to zero.

Since we had no available device to measure the density of atoms in the beam, such measurements were not performed. However, the agreement between the above equations and experiment was fully supported by the temperature dependence of ionic signals obtained in the case of three-photon ionization of potassium by the light of a ruby laser under its constant intensity. This dependence is presented in Fig. 4. The theoretical dependence was obtained on the basis of Eq. (22). The ionic signal is proportional to the density of atoms.

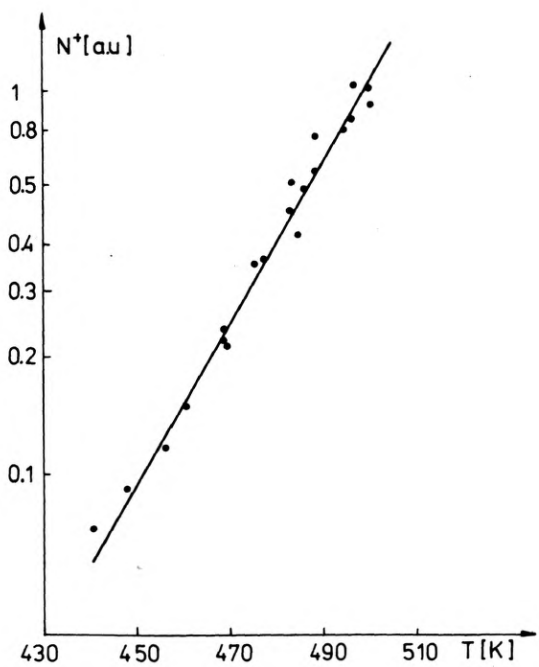

Fig. 4. Ion yield as a function of the oven temperature for three-photon ionization of potassium

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Условия конструкции двухступенчатого источника пучка щелочных атомов с мультиколлиматорным отверстием

В работе описаны условия конструкции двухступенчатого источника пучка щелочных атомов, предназначенного для исследования многофотонных процессов. Конструкция источника обеспечивает получение интенсивного пучка с малым расхождением и более низкой концентрацией димеров при небольшом использовании исследуемого металла.