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RADIOACTIVE ISOTOPE AND ISOMER SEPARATION
WITH USING LIGHT INDUCED DRIFT EFFECT

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I. Introduction

After the Light Induced Drift (LID) was theoretically predicted [1] and experimentally discovered in atomic [2] and molecular gases [3] a great number of theoretical and experimental works has been dedicated to the investigation of this effect (e.g. the references in [4]). Such interest in the LID effect is based on extended prospects of LID in scientific and practical applications. Isotope separation is one of them.

The essence of LID of absorbers (atoms or molecules) can be described as follows. Let us consider the interaction between the monochromatic laser wave traveling along the x-axis and the gas of absorbers (atoms or molecules) having only two energy levels: g - the ground level and e - the excited one. The radiation frequency is supposed to be slightly detuned from the g-e transition frequency, so that Ω is their difference. Let's consider excited and unexcited absorbers x-velocity distributions. In the absence of a laser radiation, the ground state velocity distribution is the Maxwell one, and there aren't any excited particles. In the presence of the electromagnetic wave only particles with Doppler shift compensating the detuning Ω will interact with the radiation, due to the Doppler effect. In other words, the radiation provides the transitions from the ground state to the excited one for particles with velocities about $\Omega/|\vec{k}|$, where $\vec{k}$ is the wave vector. Consequently, the ground state velocity distribution has there the hole, and, respectively, the excited state velocity distribution - the peak (FIG. 1). These are the well-known Bennet's hole and peak.

Such asymmetrical velocity distributions are equivalent to the case, when the excited particles move in the certain direction and unexcited particles move predominantly in the opposite one. These fluxes are equal, and whole gas doesn't move.
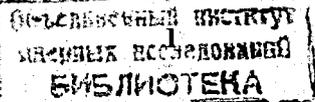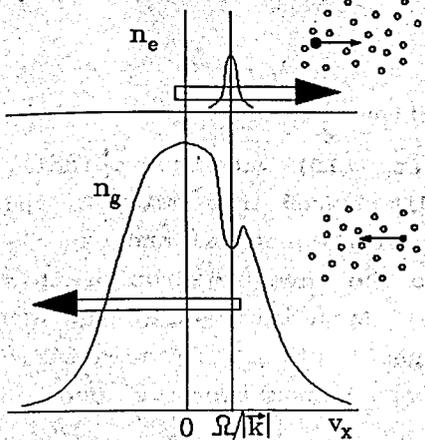
FIG. 1. *Simplified picture of LID of absorbers (•) immersed in a buffer gas (○). Velocity selective excitation of the absorbers yields asymmetric velocity distributions, both for the ground state and for the excited state absorbers. Due to the state dependence of the absorbers collision cross section, the corresponding absorber fluxes feel different collision resistances in the buffer gas; as a result a net absorber flux occurs*

The situation is quite different when these flux movements take place in another gas not interacting with the laser radiation (the so-called buffer gas). In that case the fluxes feel the collision resistance. Since gas kinetic cross sections for g- and e-states with buffer gas particles are different, the collision resistance forces for the excited and unexcited particle fluxes differ too. Consequently, the whole gas of absorbers is forced by the buffer gas. That force provides the drift of the absorber gas through the buffer one.

The LID velocity **U** is defined by

$$U \sim \bar{v} \frac{\sigma_e - \sigma_g}{\sigma_g} \varphi \, w, \tag{1}$$

where $\bar{v}$ is the average thermal absorber velocity, $\sigma_{e,g}$ are the gas kinetic cross sections for the excited and ground states absorbers in the buffer gas, w is the ratio of the excited and unexcited absorbers, φ is the factor (special for LID) which is selective according absorber velocity and antisymmetrical according laser wavelength detuning. The factor φ can be near to 1 in conditions which are optimal for LID. The exact expression for the LID velocity is deduced in [5].

We see from Eq. (1) that the drift velocity can be of the order of the thermal velocity $\bar{v}$ if large values of η and $(\sigma_e - \sigma_g)/\sigma_g$ can be realized. For Na + Xe system $(\sigma_e - \sigma_g)/\sigma_g = 0.37 \pm 0.05$ [6] and the maximum experimental LID velocity has reached about 52 m/s (10% of the thermal velocity) [7].

The optical resonance wavelengths for absorbers containing different isotopes are usually slightly shifted relative to each other. It enables to tune the laser radiation wavelength in such a way that the LID fluxes of absorbers with different isotopes have different (even opposite) velocities. This effect can produce a separation of the absorbers and, consequently, their isotopes. It has been investigated theoretically for phenomena in stellar atmospheres [8] and experimentally for some pairs of isotopes: ^{85,87}Rb with LID of atoms [9] and ^{12,13}C, ^{14,15}N, ^{79,81}Br, ^{16,18}O and ^{32,34}S with LID of molecules [10].
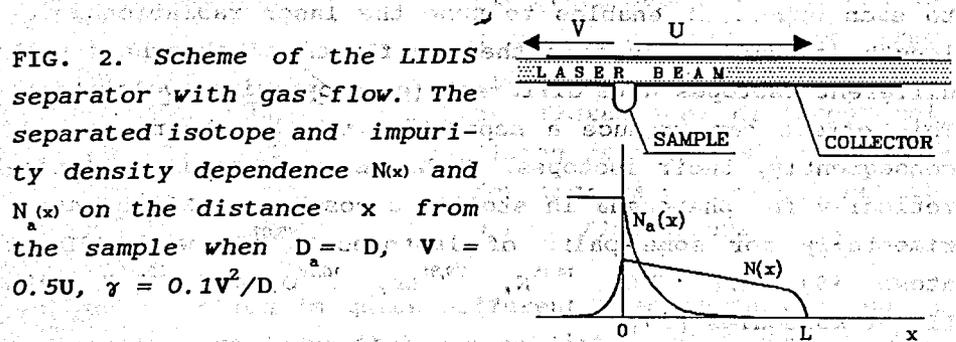
The present paper proposes the LID application for the radioactive isotope separation. In Sec. II we give the basic theory of LID separation and deduce its losses, enrichment and productivity. In Sec. III we describe the first experimental separation of the radioactive isotopes (^{22,24}Na) by using LID effect. The experimental results are discussed in Sec. IV.

II. Theory of the isotope separation with using LID

Let us call the isotope separation with using LID shortly LIDIS (**L**ight **I**nduced **D**rift **I**sotope **S**eparation). The LID of atoms would be preferred over the LID of molecules in LIDIS applications because LID effects for atoms are far stronger than for molecules. Therefore, below LID is considered as LID of atoms only (not molecules).

The principal characteristics of LIDIS are deduced for the separator illustrated schematically in FIG. 2. The main part of the separator is a separative pipe with a little inner diameter (1 ÷ 5 mm). The sample with an isotope mixture is located in an appendix. An inert gas flow with velocity **V**

and a laser beam pass through the separative pipe. Atoms of the isotopes are released by evaporation or in any other way and diffuse into the separative pipe. In an ideal case the laser radiation interacts only with atoms of one of the sample isotopes and produces LID of these atoms in the opposite direction to the gas flow and with the velocity $U \approx 2V$. Only the separated isotope atoms reach a collector and the remainder are blown off by the gas flow.

FIG. 2. *Scheme of the LIDIS separator with gas flow. The separated isotope and impurity density dependence* $N(x)$ *and* $N_a(x)$ *on the distance* x *from the sample when* $D = D_a$, $V = 0.5U$, $\gamma = 0.1V^2/D$

The separated isotope atom density N is described by this diffusion equation:

$$Dd^2N/dx^2 + (U - V)dN/dx - \gamma N = 0, \qquad (2)$$

where D is a diffusion coefficient and γ is the coefficient of the losses due to chemical bounding. If the boundary conditions are $N(0) = N_0$ and $N(L) = 0$ (L is the distance from appendix to collector) the separated isotope atom density is given by:

$$N(x) = N_0 \frac{\exp[\alpha_1(L - x)] - \exp[\alpha_2(L - x)]}{\exp(\alpha_1 L) - \exp(\alpha_2 L)}, \qquad (3)$$

$$\alpha_{1,2} = (V - U)/(2D) \pm \{[(V - U)/(2D)]^2 + \gamma/D\}^{1/2}.$$

If $\gamma \ll (U - V)^2/4D$ and $L(U - V)/D \gg 1$ are hold then

$$N(x) = N_0 \left\{ \exp\left[-\frac{\gamma x}{U - V}\right] - \exp\left[-\frac{\gamma L}{U - V} - \frac{U - V}{D}(L - x)\right] \right\} \qquad (4)$$

An example of the dependence $N(x)$ is given in FIG. 2. The flux j of the separated isotope atoms on the collector is determined by

$$j = -D\nabla N + (U - V)N|_{x = L} \approx (U - V)N_0\exp[-\gamma L/(U - V)]. \qquad (5)$$

If $\gamma \ll (U - V)/L$ is true, the great part of separated isotope will reach the collector.

For impurity isotopes the diffusion equation (2) is true if $U = 0$ and $\gamma = 0$. Under similar boundary conditions the impurity isotope density $N_a(x)$ is defined by

$$N_a(x) = N_{a0}[\exp(-Vx/D_a) - \exp(-VL/D_a)][1 - \exp(-V/D_a)]^{-1}. \qquad (6)$$

If $\exp(-VL/D_a) \ll 1$ is held, then

$$N_a(x) = N_{a0}\exp(-Vx/D_a), \qquad j_a = VN_{a0}\exp(-VL/D_a). \qquad (7)$$

An example of the dependence $N_a(x)$ is given in FIG. 2.

The separated isotope enrichment E on the collector can be expressed by

$$E = (U - V)/V\exp\{L[V/D_a - \gamma/(U - V)]\} - 1. \qquad (8)$$

As $\gamma \ll (U - V)/L$ and $U = 2V$ are held, the expression (8) can be simplified:

$$E \approx \exp(LV/D_a). \qquad (9)$$

The LID velocity $U \approx 5$ m/s for sodium (the most investigated element) atoms can be easily reached at diffusion coefficient $D \approx D_a \approx 20$ cm^2/s [6]. If the buffer gas flow velocity $V \approx U/2 \approx 2.5$ m/s, then the parameter D_a/V in (9) can be less than 0.1 cm and the enrichment E can be higher than $\exp(10) \approx 2.2 * 10^4$. There is no principle obstacle to reach arbitrary high enrichment.

The productivity of the method is limited by laser light absorption in the separating pipe. Suppose that the laser light is absorbed by the separated atoms only. For optimum density of the separated atoms the following condition is fulfilled:

$$\alpha L \sim 1, \qquad (10)$$

where α is the absorption coefficient that can be determine

from photon density Φ equation:

$$d\Phi/dx = -\alpha\Phi = -Np, \qquad (11)$$

where p is the absorption frequency of photons by one atom. Using (9), (10) and (11) we can determine maximum density N_{max} of separated isotope atoms:

$$N_{max} \sim \Phi/(Lp) = \Phi V/(D_a p*lnE). \qquad (12)$$

The maximum flux j_{max} of separated isotope atoms can be got by substitution of (12) into (5) when $\gamma L/(U - V) \ll 1$ is held:

$$j_{max} \sim (U - V)V\Phi/(D_a p*lnE). \qquad (13)$$

Substitution of $U \approx 2V$ and $D_a \approx D = \bar{v}^2/(2\nu)$ ($\bar{v}$ is thermal velocity and ν is the effective collision frequency) into (13) yields

$$j_{max} \sim \Phi/(2*lnE)*(U/\bar{v})^2*(\nu/p). \qquad (14)$$

Substitution of $\nu/p \sim 1$ (optimum condition for LID [5]), $\Phi \sim 10^{19}$ photon/s ($\sim$ 1 Watt laser radiation power in visible region), $U \sim 10$ m/s, $lnE \approx 10$, and $\bar{v} \sim 500$ m/s into (14) gives

$$j_{max} \sim 10^{14} \text{ atom/s}.$$

It is worth noting that for isotope with mass number about 100 the separation productivity about 1 μg/hour corresponds with the flux $\sim 10^{12}$ atom/s. It can be reached with LID velocity $U \sim 1$ m/s.

III. Experimental method

We selected sodium as an object for the first LIDIS experiment owing to the following reasons. At first, the LID of stable sodium atoms is well investigated [6], the optimum experimental conditions and the corresponding LID velocity are known. At second, ^{22}Na and ^{24}Na are suitable isotopes for the LIDIS investigations. Their isotope shifts for the D_1 line are -756.9 and 706 MHz, correspondingly [11] (FIG. 3). The laser radiation wavelength tuning at the center of the ^{23}Na line produces the LID of ^{22}Na and ^{24}Na atoms in opposite directions. Both LID velocities of the ^{22}Na and ^{24}Na atoms are near the maximum velocity that can be reached at the optimum laser wavelength tuning [12, 13]. That is why we used the separating device without the buffer gas flow.
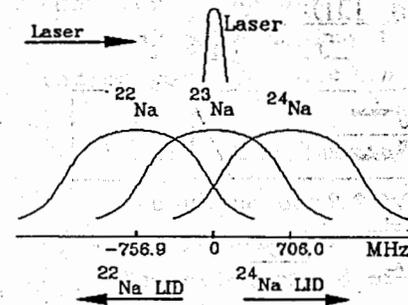

FIG. 3. *Illustrative picture of the tuning of the laser radiation wavelength at D_1 line of atoms of the sodium isotopes: The curves represent the approximate spectral contours of the laser line and Doppler broadened lines of the sodium isotopes. The arrows represent the directions of the laser radiation propagation and LID of ^{22}Na and ^{24}Na atoms*

A scheme of the experimental set-up is given in FIG. 4. The center of a stainless separating pipe 1 with the inner diameter of 1.2 mm and the length of 10 cm was connected with a stainless appendix 2. In the appendix 2 there was a graphite boat 3 with a metal aluminium 4 (100 mg) on it. The aluminium 4 contains about 10^{12} atoms of ^{22}Na ($T_{1/2}$ = 2.62 y) and about 10^9 atoms of ^{24}Na ($T_{1/2}$ = 14.96 h). The isotopes had been produced by the aluminium spallation on 660 MeV beam. At the ends of the separating pipe 1 there were two collectors 5 made of quartz. It binds well sodium atoms at high temperatures. The separating pipe 1 is fixed in a quartz jacket 6. The jacket contained 5 torr of krypton. The reactive impurity in the buffer gas which could bind sodium atoms (O_2, H_2O and so on) was eliminated by potassium vapor. The separating pipe 1 and the appendix 2 had been baked out for 3 h at 1200 K and 850 K, respectively, in vacuum before the gas filling of the jacket 6.

The beam of a cw ring dye laser (Spectra Physics 380 D) with a power of about 40 mW passed through the separating pipe 1. The laser radiation wavelength was tuned at the D_2

line center of the ^{23}Na atomic spectrum (λ = 588.99 nm). (LID effect of sodium atoms is stronger for D_2 line then for D_1 one.) We suppose that isotope shifts in the D_2 line are approximately the same as for the D_1 line.
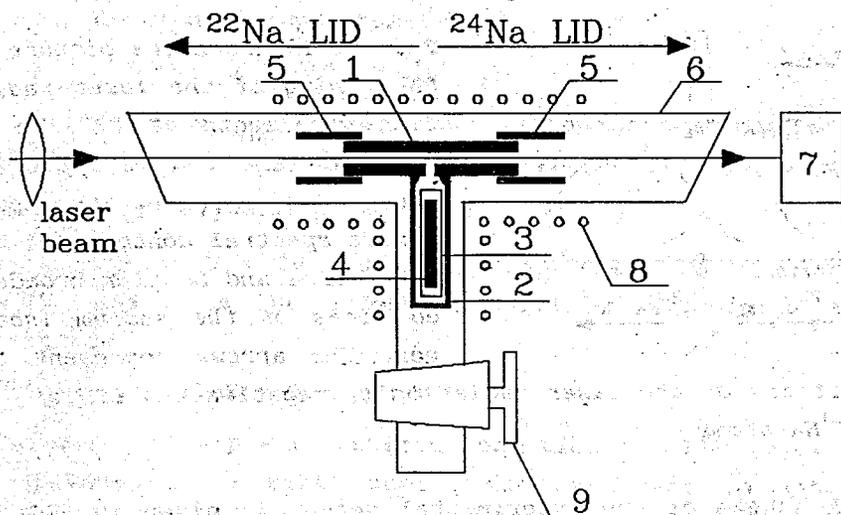

FIG. 4. *Scheme of the LIDIS separator. The figures on the picture represent: 1 - separating pipe, 2 - appendix, 3 - graphite boat, 4 - aluminum sample, 5 - collectors, 6 - jacket, 7 - power meter, 8 - heater, 9 - valve. The arrows represent the directions of LID of ^{22}Na and ^{24}Na atoms*

The separating pipe 1 was heated up at 1200 K. This temperature is above the adsorption temperature of sodium atoms and molecules of sodium compounds on a stainless steal surface. The appendix 2 was heated up above the aluminium melting point (933 K). The sodium atoms diffused in the liquid aluminium to its surface. They were evaporated there and diffused in gas phase into the separating pipe 1. LID swept the ^{22}Na and ^{24}Na atoms in the separating pipe 1 to the left and right, respectively. It produced ^{22}Na/^{24}Na enrichment on the left/right collectors 5. A power meter 7 measured the power of the laser beam that transmitted through the separating pipe 1. The sodium atoms density in the separating pipe 1 was kept in an optical thin range by a control of the

appendix 2 temperature. The power of the laser beam passed through the separating pipe was kept above 20 mW. We suppose that the laser beam was predominantly absorbed and attenuated by the ^{23}Na atoms which had been contained in larger amount than the ^{22,24}Na atoms in the aluminium sample 4. The separation was made during 1 hour. Then the ^{22}Na and ^{24}Na activity ratios were measured on the collectors 5.

IV. Experimental results and their discussion

Let us compare the theoretical and real characteristics of the separator at the conditions of this experiment.

An enrichment E of ^{22}Na can be expressed as

$$E = \frac{A_k^{22} A_s^{24}}{A_k^{24} A_s^{22}} - 1, \qquad (15)$$

where A_s^I and A_k^I are indicated isotopes activities in the sample and on a collector respectively at a certain time. In our experiment the ^{22}Na enrichments E^{ex} were $(11 \pm 4) \cdot 10^{-2}$ and $(-19 \pm 2.7) \cdot 10^{-2}$ for left and right collectors, respectively (FIG. 5).

The theoretical enrichment E^{th} of ^{22}Na on the left collector can be determined by

$$E^{th} = \exp(Lu^{24}/D^{24}) - 1, \qquad (16)$$

where L is the distance between the center of the separating pipe 1 and the left collector, u^{24} and D^{24} are LID velocity and diffusion coefficient for ^{24}Na atoms. The experimental values of the quantities were L = 5 cm, $u^{24} \approx$ 200 cm/s, $D^{24} \approx$ 200 cm^2/s [6] (u^I and D^I are only estimated). Their substitution into (16) gives $E^{th} \sim 100$.

The real experimental value of the enrichment E^{ex} is considerably lower than the theoretical one. This discrepancy can be accounted if we suppose the main part of sodium was chemically bound with gas impurities into molecules and free sodium atoms consisted only of a little part of all the sodi-

um. In that case the isotope enrichment considerably depended on a ratio of the densities of the sodium atoms and molecules in gas phase.

FIG. 5. *The ^{22}Na and ^{24}Na activity ratios A^{22}/A^{24} and the ^{22}Na enrichments E in the primary sample 4 and on the collectors 5 after the experiment. The arrows represent the directions of LID of ^{22}Na and ^{24}Na atoms*
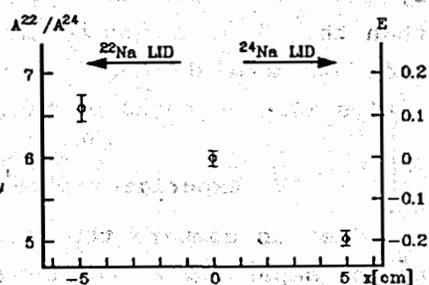
The sodium isotope enrichment can be increased by using the following ways: 1) A buffer gas purification from impurities which can bound the sodium atoms. 2) A buffer gas flow through the separating pipe 1. 3) A separating pipe heating above the decomposition temperature of molecules with sodium atoms. 4) A molecule decomposition by an electron bombardment in the separating pipe 1. The last two ways are very perspective because they can give a possibility to use compounds as a sample of isotopes for separation and to separate isotopes of little volatile elements.

V. Conclusions

The new gas-kinetic phenomenon called Light Induced Drift (LID) has a large application prospect in the separation of a microgram amount of radioisotopes. The new laser isotope separation method called LIDIS (Light Induced Drift Isotope Separation) can produce an isotope separation simultaneously with both a high enrichment and a low losses. The LIDIS productivity can reach theoretically the value of 10^{14} atom/s.

The first laser separator with using LID has been built up and tested. In its first experiment it has separated radioactive sodium isotopes ^{22}Na and ^{24}Na. The separator cha-

racteristics were influenced considerable by a stable sodium content in the sample and by an impurity content in the buffer gas. In the separation experiment the isotope enrichment was 0.15 (theoretical value more than 100). In future experiments we plan to increase the enrichment and the productivity and to reduce the losses of the separation at the theoretical levels by a buffer gas cleaning and an improvement of the LIDIS separator construction.

LIDIS should become more universal and technological method if molecules will be decomposed in separating pipe. Such conditions would be a high temperature (up to 3300 K) or an electron bombardment.

LIDIS should bring about price decrease of radioisotope application in medicine (e. g. In, Tl and so on) and in science (e. g. in nuclear physics experiments with ^{178m}Hf). We investigate the ways of overcoming these problems.

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