Laser light-induced drift of radioactive ^{22}Na and ^{24}Na

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Laser light-induced drift of ^{22}Na and ^{24}Na in Kr as the buffer gas is investigated. The resonance frequency at which the ^{22}Na and ^{24}Na atoms drift in opposite directions is determined for the D_2 line. The distribution of both isotopes along the drift tube was measured at 300 K and 1000 K. These measurements were performed by recording the γ-rays emitted as the isotopes decay. An isotope separation factor of up to 20 with an efficiency of 50% and drift velocity of 1.1 m/s was achieved.

1. INTRODUCTION

Light-induced drift (LID) of atoms in a buffer gas is one of the effects observed when laser radiation interacts with matter.[1,3] This effect appears when the atoms of a selected isotope are excited selectively, according to their velocity, by laser radiation. The frequency of the excitation line, which is narrower than the Doppler profile, is chosen so that only atoms moving with a definite velocity in a prescribed direction relative to the laser beam are excited. As a rule, excited atoms are bigger than ground-state atoms, and for this reason their collision cross sections with the buffer gas is larger and their diffusion coefficients are smaller. As a result, oppositely directed fluxes of excited and unexcited atoms appear, changing the concentration of the atoms. Sufficiently far from the source of atoms, this change can be considerable (by factors of tens and hundreds). This effect could find many applications in different fields of physics and technology.

Light-induced drift has thus far been observed for a comparatively small number of elements (for example, Na,[2,3] Rb,[4] and Ne[5]) as well as for a number of molecules.[6] A detailed theoretical description of this effect for molecules is given in Ref. 7. Extensive investigations of this phenomenon have been performed for Na atoms.[8,9] However, many of these investigations were performed at close to room temperature in a narrow capillary which was either made of special glass or whose inner walls were coated with a paraffin film or a film of diffusion-pump oil in order to prevent adsorption of Na vapor. At the same time, it is obvious that the possible diverse applications of LID might require different conditions, for example, much higher temperatures, capillaries made of other materials, and so on. All this makes it interesting to perform further investigations of LID of Na atoms and to enlarge the range of elements and isotopes studied.

Investigations of LID will be more effective if radioactive atoms are used instead of stable atoms, as done in all preceding experiments. The sensitivity of the measurements can be increased considerably by recording, instead of the resonantly scattered optical radiation, the γ-rays emitted during the decay of radioactive isotopes. This technique also makes it possible to determine the concentration of each experimental isotope both during drift and integrated over the entire time of the experiment. All these measurements can be performed using different isotopes in the same experiment, thereby expanding considerably the possibilities of LID investigations.

Our objective in the present work is to investigate LID of radioactive ^{22}Na and ^{24}Na under new conditions—at higher temperatures and in drift tubes made of more heat-resistant materials (stainless steel, quartz). These investigations include measurements of both the dynamic characteristics (changes in the Na concentration in time at the scanned and fixed laser frequency) and the steady-state characteristics (concentration distribution along the tube during drift and integrated over the entire time of the experiment).

2. THEORY

Light-induced drift has been investigated in detail both theoretically and experimentally.[8,9] If the source of atoms is located at the center of the drift tube ($x=0$), then the time-dependence of the concentration $N(x,t)$ of atoms in different sections of the tube in the presence of drift with velocity u can be described by the one-dimensional diffusion equation

$$dN/dt-\mathrm{div}(uN-\nabla DN)+\gamma N=0, \qquad (1)$$

where D is the effective diffusion coefficient and γ is a constant characterizing the loss of the experimental atoms as they move along the tube (γ is the inverse average lifetime of the atoms in the buffer gas and depends strongly on the quantity and composition of impurities reacting with the free Na atoms).

The solution of Eq. (1) shows how LID changes the concentration of the experimental atoms in different sections of the drift tube under different experimental conditions. For example, the steady-state concentration distribution (which is ordinarily determined in an experiment) is described by the following expressions:

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in the drift direction ($x<0$):

$$N(x)=N_0\exp(x/l_1),\quad l_1=\left[\left(\frac{u^2}{4D^2}+\frac{\gamma}{D}\right)^{1/2}+\frac{u}{2D}\right]^{-1}\tag{2}$$

opposite the drift ($x>0$):

$$N(x)=N_0\exp(x/l_2),\quad l_2=\left[\left(\frac{u^2}{4D^2}+\frac{\gamma}{D}\right)^{1/2}-\frac{u}{2D}\right]^{-1}\tag{3}$$

in the absence of drift ($u=0$), when only diffusion of the atoms occurs:

$$N(x)=N_0\exp(x/l_0),\quad l_0=(D/\gamma)^{1/2}.\tag{4}$$

It is evident from the expressions (2)–(3) that LID produces an asymmetric concentration distribution of the atoms with respect to their source. The concentration ratio (the LID asymmetry factor) at the same distance from the source in the drift direction and in the direction opposite the drift increases exponentially with x:

$$\beta(x)=\exp\frac{u}{D}x.\tag{5}$$

The growth rate of $\beta(x)$ is determined by the ratio of the LID velocity to the diffusion coefficient of the experimental atoms in the buffer gas. The expression for the LID velocity contains a product of three quantities:

$$u=\frac{D_g-D_e}{D_g}\eta V_L.\tag{6}$$

The first factor determines the relative change of the diffusion coefficient as a result of a transition from the ground state (D_g) into an excited state (D_e). In the diffusion of Na atoms in an inert gas, this change ranges from 0.14 for He to 0.49 for Xe.[9] The second (η) and third (V_L) factors are, respectively, the fraction of atoms in the Doppler profile that are excited by laser radiation at a fixed frequency and the average projection of their thermal velocity on the drift direction. The product of these factors within the Doppler profile is a complicated function of the laser frequency. It vanishes at a frequency corresponding to the center of the profile (atoms moving perpendicular to the laser beam are excited) and reaches a maximum when the frequency is shifted by

$$\Delta\nu=\frac{v}{c}\nu\tag{7}$$

(ν is the resonance frequency and c is the speed of light) from the profile center; atoms whose most probable thermal velocity v is parallel or antiparallel to the laser beam are then excited.

Accordingly, at two frequencies, shifted by the amount $\Delta\nu$ to the left or right of the profile center, the LID velocity is maximum and oppositely directed. The buffer gas, the characteristics of the optical transitions, and the parameters of the laser radiation determine the maximum LID velocity. Under optimal conditions, drift velocities $u>10$ m/s can be achieved. In this case $u/D>10$ cm^{-1}, and appreciable asymmetry factors are found even several millimeters from the source.

TABLE I. Parameters of the optical spectra of Na isotopes.

Isotope	$\Delta\nu_{is}^{23,A}$, MHz	$\Delta\nu^{F,F'}$, MHz	$\Delta\nu_d$, MHz	
			300 K	1000 K
^{22}Na	-750	1240	1345	2470
^{23}Na	0	1777	1315	2410
^{24}Na	+706	1140	1290	2360

This picture is correct, however, only in a simple two-level scheme (ground state and an excited state). In Na atoms, splitting of both the $s_{1/2}$ ground state and the $p_{1/2}$ and $p_{3/2}$ excited states is observed, and the ground-state splitting is much larger than the splitting in excited states. In this level scheme, the excitation of the different components of the levels can result in drift in opposite directions. During LID, collisions with buffer-gas atoms will induce transitions between the components of the excited states, optical pumping on one component of the ground state will occur, and interactions with the walls of the drift tube will induce transitions to a different component. All this can change the drift velocity (6) considerably. This change in the LID velocity is determined by the optical characteristics of the atomic levels of the experimental isotopes (isotopic shift $\Delta\nu_{is}$ and hyperfine splitting $\Delta\nu^{FF'}$ of the ground state) and by the broadening of the spectral lines (Doppler broadening $\Delta\nu_d$ and collisional broadening $\Delta_{\text{int}}\sim200$ MHz). These quantities for ^{22}Na, ^{23}Na, and ^{24}Na under our experimental conditions, which are described below, are presented in Table I. It is obvious that some of these quantities are somewhat different for different Na isotopes.[10] This is why investigations of LID using different Na isotopes make it possible to determine how these parameters affect the velocity and other characteristics of drift.

3. EXPERIMENTAL SETUP

The experimental setup for investigating LID of Na isotopes consisted of a laser spectrometer, a collection of stainless steel drift tubes of different length and diameter, and systems for evacuating the tubes and filling them with inert gas (Fig. 1).

The laser spectrometer consisted of a model SP 280D dye laser pumped with an SP 2030 cw argon-ion laser. The laser frequency could be scanned over a selected range up to 30 GHz wide. A stabilization system made it possible to maintain the chosen frequency to within 10–20 MHz. A Burleigh WA-10 wavemeter and a Na-vapor lamp were used to tune the laser over the prescribed frequency range. An OPHIR 20A power meter was used to monitor the intensity of the laser radiation transmitted through the drift tube.

Light-induced drift of Na atoms occurred in the stainless steel tube. Tubes of different diameter (ranging from 1.2 to 4.0 mm) and up to 60 mm long were used. A branch piece holding a volatile sample containing the experimental atoms was attached at the center of the tube. The tube, together with the branch piece, was placed in a quartz cell, which was heated to the prescribed temperature with a nichrome coil. The branch piece with the sample could also be heated inde-

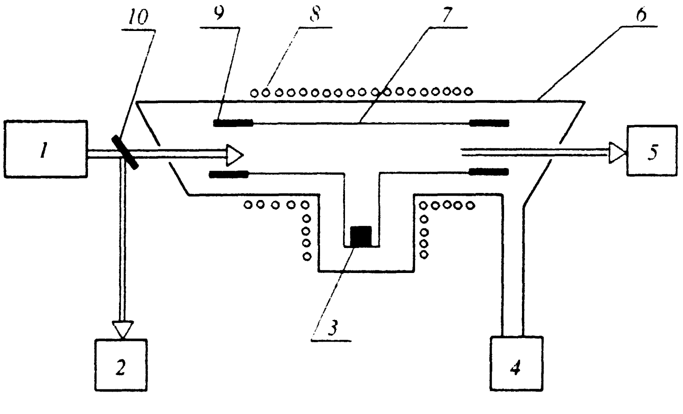

FIG. 1. Block diagram of the experimental setup. *1*—Laser spectrometer; *2*—wavemeter; *3*—sample with ^{22}Na and ^{24}Na; *4*—evacuation system; *5*—radiation power meter; *6*—vacuum chamber; *7*—drift tube; *8*—heater; *9*—atom collector; *10*—mirror.

pendently. A chromel–alumel thermocouple and a pyrometer were used to monitor the temperature of the cell and the drift tube.

Roughing and diffusion pumps were used to produce a vacuum of 10^{-4} torr in the cell. Special care was taken to eliminate impurities that would bind Na atoms as a result of chemical reactions (water vapor, oil vapor, oxygen, and hydrogen). The sodium compounds formed participate only in diffusion, and not in drift, and they therefore make drift less efficient. For this reason, during evacuation the quartz tube was heated at a temperature of 850 K for several hours. Potassium and zirconium, heated up to this temperature, were used to absorb the impurities released when the quartz enclosure and the drift tube were heated. The inert gas filling the cell was passed through a liquid-nitrogen trap.

In the room-temperature experiments, an aluminum foil placed inside the drift tube was used to collect the ^{22}Na and ^{24}Na atoms. In experiments at high temperatures, quartz collectors at both ends of the tube were used (quartz binds Na atoms well at high temperatures).

In previous papers, the individual elements of the experimental apparatus, the experimental procedure, and some experimental results are described and the results of the analysis are described in greater detail.[11–13]

The decay characteristics of the isotopes ^{22}Na and ^{24}Na used in these experiments were considerably different: ^{22}Na has a half-life of 2.6 yr and it emits 511 keV and 1274 keV γ-rays, and ^{24}Na has a half-life of 15.0 h and emits 1368 keV and 2.754 keV γ-rays.[14] This enabled us to distinguish clearly, using γ-ray spectrometers (a scintillation spectrometer with a NaI(Tl) crystal or a semiconductor spectrometer with a Ge(Li) crystal), the radiation of each isotope, to measure the concentration of the isotopes in the drift tube, and to perform LID experiments using both isotopes simultaneously.

The isotopes ^{22}Na and ^{24}Na were produced in a spallation reaction by irradiating ultrapure Al foil with 660 MeV protons and the ^{24}Na isotope was also produced in the reaction ^{27}Al(n,α) with uranium-fission neutrons.

4. EXPERIMENTAL RESULTS

Two types of experiments on light-induced drift of the radioactive isotopes ^{22}Na and ^{24}Na were performed using the setup described above.

1. The ^{24}Na concentration at the end of the drift tube was measured as a function of the laser frequency and as a function of time with the laser turned on and off. The purpose of these measurements was to study the interaction of Na atoms with the hot wall of the tube and to determine the laser frequency at which the LID velocity is maximum.

The experiments were performed in a 2.5-mm diameter and 150-mm long stainless steel drift tube, filled with Kr to a pressure of 10 torr. One end of the tube was sealed, and the ^{24}Na concentration was measured at this end according to the intensity of the γ-radiation. A NaI(Tl) scintillation spectrometer was used for these measurements. The spectrometer was shielded with a 50-mm thick lead shield from the $1.2\cdot10^5$ Bq ^{24}Na source in the branch piece. The source was ultrapure Al foil irradiated by protons.

When the drift tube and the branch piece containing the ^{24}Na source were heated to 1000 K and the tube was filled with Na vapor as a result of diffusion, the intensity of the γ-radiation from the sodium increased. When the γ-ray intensity approached saturation (indicating almost complete vaporization of the ^{24}Na contained in the sample), the laser frequency was scanned in the neighborhood of the D_2 line of sodium ($\lambda=589.0$ nm) at a rate of 1 GHz/min. The laser radiation directed from the left end of the tube filled the entire area of the drift tube; the average power density of the radiation was 0.6 W/cm^2. A maximum at a frequency close to the resonance frequency for the ^{23}Na isotope is clearly visible in the intensity of the γ-radiation from ^{24}Na measured as a function of the laser frequency (Fig. 2). This maximum obviously corresponds to the highest LID velocity in the direction of the laser beam. The minimum associated with drift in the opposite direction is much less pronounced because of the background produced by ^{24}Na atoms adsorbed on the walls of the tube.

Measurements of the time dependence of the γ-ray intensity (or the ^{24}Na concentration) were performed at this frequency with the laser radiation turned off and on. When the laser was turned off, the ^{24}Na concentration at the end of the drift tube dropped by a factor of two within 20 s. When the laser was turned on, the ^{24}Na concentration was restored at the same rate to the previous level. These changes in the ^{24}Na concentration are much slower than those associated with the diffusion of Na atoms in Kr at fixed pressure and temperature (in this case, according to the calculations, the

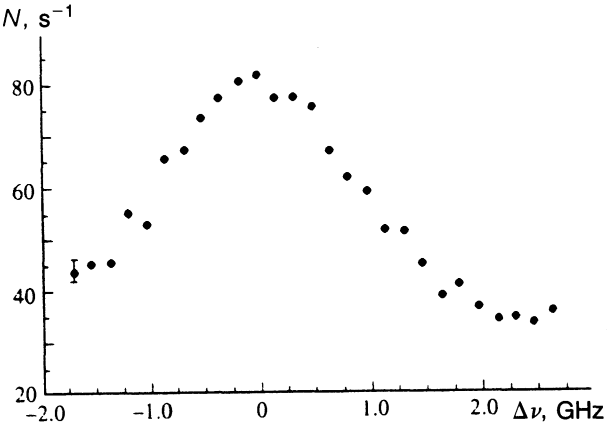

FIG. 2. γ-ray counting rate versus laser wavelength. The frequency shift $\Delta\nu=0$ corresponds to the center of the D_2 line of the ^{23}Na atoms.

concentration would change by a factor of two within ~1 s). This large difference is explained by absorption and subsequent desorption of ^{24}Na atoms at the walls as the atoms move along the drift tube. In the process, the ^{24}Na atoms spend ~95% of their time in the tube at the walls. This time obviously depends on the material and temperature of the tube. For example, when the steel tube was replaced by a copper tube, the ^{24}Na concentration changed much more slowly (the concentration dropped by a factor of two in ~4 min).

Figure 3 shows the arrangement of the Doppler profiles vs. frequency of the hyperfine components of the optical D_2 line of the atoms of the experimental Na isotopes (the hyperfine splitting of the excited $p_{3/2}$ level was neglected; it is significantly smaller than the splitting of the ground state). As mentioned above, the maximum LID velocity is observed at a laser frequency corresponding to the center of the Doppler profile of ^{23}Na atoms. It is obvious from Fig. 3 that the ^{24}Na and ^{22}Na hyperfine component with total spin $F=9/2$ and 5/2, respectively, lies near this frequency (within the homogeneously broadened excitation line), and the other components are shifted to a frequency close to that required to achieve the maximum LID velocity. At this frequency the influence of optical pumping (which is not favorable for LID) of the component is much weaker.

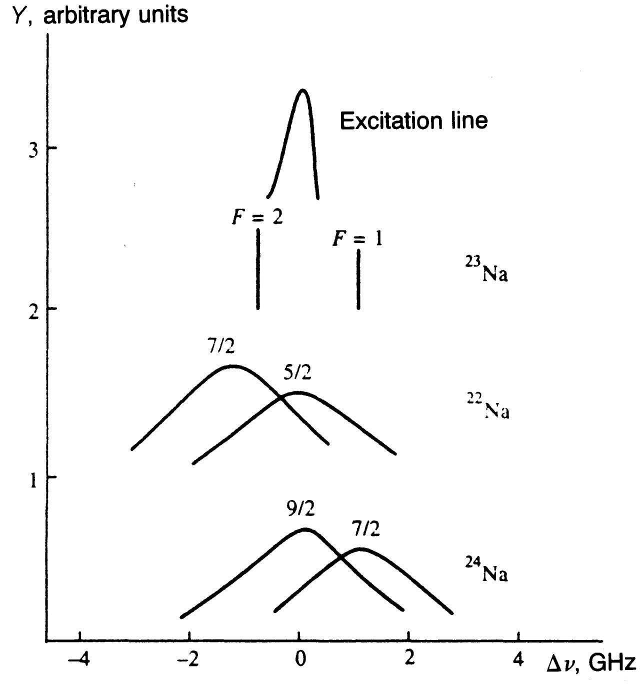

FIG. 3. Excitation line and Doppler profiles of the hyperfine components of the optical D_2 line of ^{22}Na and ^{24}Na.

The width (~2 GHz) of the observed resonance is close to that of the Doppler profile for a single component (Table I) at $T=1000$ K.

2. The distribution of the ^{22}Na and ^{24}Na concentrations along the drift tube with the laser frequency corresponding to the center of the Doppler profile of ^{23}Na was measured. As one can see from Fig. 3, at this frequency ^{24}Na and ^{22}Na drift in opposite directions: parallel and antiparallel, respectively, to the laser beam. Measurements of the spatial distribution of each isotope in the same experiment enable one to make a more definitive determination of the LID velocity and its dependence on different factors.

The experiments were performed at room temperature ($T=300$ K) in a wide (10-mm diameter) drift tube, and at $T=1000$ K in a 3-mm diameter tube. This corresponded to completely different conditions of LID. In the first case the Na atoms were absorbed by cold walls and the Na concentration decreased exponentially along the tube. In the second case the Na atoms were desorbed from the hot walls and drift occurred with essentially no losses.

Two types of sources of radioactive Na isotopes were used in the experiments: one source was aluminum foil and the other a graphite rod, which, when heated to 1000 K, absorbed ^{22}Na and ^{24}Na in vacuum. The first source was heated to a temperature above the melting point of Al (933 K), and the second was heated to 2100 K. When the sources were heated, sodium was liberated, in spite of the fact that materials with a high degree of purity were used for the sources (this was made noticeable by the decrease in the laser intensity transmitted through the drift tube). The surface of the sources was probably contaminated with sodium. The sources were therefore preheated for 10–15 s to eliminate contaminants; over this period of time there was virtually no decrease in the radioactivity of the Na isotopes from the volume of the sources. The Na isotopes were completely evaporated within 15–30 min, and laser radiation with frequency corresponding to resonance with the ^{23}Na D_2 line (resonance was monitored with a wavemeter and a sodium vapor lamp) was passed through the tube from the left end throughout this entire time.

After evaporation ceased, a Ge(Li) spectrometer was used to measure the intensity of the γ-radiation from the ^{22}Na and ^{24}Na isotopes in different sections of the drift tube to the left and right of the source. Usually, ~50% of the radioactive atoms remained on the walls of the branch piece where the sources were located, and the rest of the atoms entered the tube and participated in drift. The ^{22}Na and ^{24}Na concentrations could be determined unequivocally by com-

TABLE II. ^{22}Na and ^{24}Na concentration ratios.

l, mm	W, W/cm^2	T, K	p, Torr	ε, %	$\alpha(l)$ left	$\alpha(l)$ right
6	0.25	300	10	3	2.8(1)	0.60(2)
6	0.20	300	3	2	3.0(1)	0.50(2)
14	0.25	300	10	0.04	9(3)	0.4(1)
20	0.20	1000	10	50	18(2)	0.20(2)
50	0.30	1000	10	50	>20	0.04(1)

paring the output of 1368 keV (^{24}Na) and 1274 keV (^{22}Na) γ-rays in the original sources and at the collectors in the drift tube.

The results of these measurements are presented in Table II, which shows the fraction (ε) of the atoms reaching the collector and the ratio $\alpha(l)$ of the ^{22}Na and ^{24}Na concentrations at different distances to the left and right of the source (normalized to the concentration ratio at the source). The experimental conditions are also presented: the pressure p, the temperature T, and the laser power density W. It is obvious that LID is observed clearly under any experimental conditions—^{24}Na enrichment along the laser beam (to the right of the source) and ^{22}Na enrichment in the direction opposite the laser beam (to the left of the source). In addition, the enrichment increases away from the source.

It is also evident from Table II that the increase in the enrichment is different on the left- and right-hand sides of the tube. At the same distances from the source α (left)$>\alpha^{-1}$ (right), i.e., enrichment grows more rapidly in the case of isotope drift opposite the laser beam than in the case of isotope drift parallel to the laser beam. There are two reasons for this difference:

1. As one can see from Fig. 3, at a chosen laser frequency the ^{22}Na and ^{24}Na atoms are excited from different hyperfine components—from the state with lower total angular momentum ($F=5/2$) for the first isotope and with higher angular momentum ($F=9/2$) for the second isotope. This may result in different values of the drift velocity.

2. Since vapor of the stable isotope ^{23}Na is present in the tube, the laser radiation is attenuated as it propagates through the tube. The drift velocity on the right-hand side of the tube is therefore lower than on the left-hand side, resulting in different degrees of enrichment.

Since the ^{22}Na and ^{24}Na atoms drift in opposite directions, it can be assumed that their concentration ratios $\alpha(l)$, presented in Table II, are approximately determined by the expression (5) with the LID asymmetry ratios

$$\alpha(l)\approx\exp\frac{\bar{u}}{D}\,l, \tag{8}$$

where $\bar{u}$ is the average drift velocity for both isotopes.

The average drift velocities $\bar{u}$ can be estimated using the expression (8), the measured values of $\alpha(l)$, and the known diffusion coefficients of Na atoms in Kr.[8] They were found to be close in magnitude for different values of l: ~0.2 m/s on the left-hand side, where the drift velocity is highest in the cold tube, and ~1.1 m/s in the hot tube. The drift velocities are two times lower on the right-hand side of the tube. It is interesting to compare these values of the drift velocity to the analogous velocities obtained in Refs. 8 and 9 for the stable isotope ^{23}Na by different methods (measurement of the asymmetry ratio or direct observation of the motion of the fluorescing volume of Na atoms). It was found that if the values obtained for the drift velocity in our experiments and in Refs. 8 and 9 are reduced to the same conditions (temperature, pressure, laser power density, shift of the laser frequency), then they are close in magnitude. Obviously, the differences in the hyperfine splittings of the Na isotopes (Table I) have little effect on the LID velocity.

5. CONCLUSIONS

The experiments performed in this work made it possible to determine the characteristics of LID of the radioactive isotopes ^{22}Na and ^{24}Na—the laser frequency at which the drift velocity is maximum, and the velocities under different conditions. These values show that agreement with the theory of LID obtains even at high temperatures (1000 K). It was also shown that high LID velocities—up to 1 m/sec—can be achieved with comparatively low laser powers (~0.2 W/cm^2). Such drift velocities make it possible to efficiently influence the motion of laser-excited atoms in a buffer gas. For example, atoms of a selected isotope can be pushed rapidly in a prescribed direction. This could find many diverse applications in laser spectroscopy, isotope separation, and so on.

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