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LASER SPECTROSCOPY ON THE BEAMS OF ACCELERATED RADIOACTIVE NUCLEI

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INTRODUCTION. High resolution laser methods of the nuclear structure investigations have been extensively developed during the past several years. Measurements of the isotopic shifts and the hyperfine splitting in the optical spectra of atoms or ions by means of these methods make it possible to determine the charge radii difference $\Delta\langle r^2\rangle$ and multipole nuclear moments (magnetic dipole μ and electric quadrupole Q). Laser methods are the most sensitive ones, they allow one to carry out the experiments with the nuclei produced in the very low yield reactions (high spin isomers, nuclei on the boundary of the nucleon stability).

As a rule, these nuclei are the short-lived ones and fast-acting experimental set-ups are required for their study. On-line mass-separators with an ion source are not suitable for a wide number of elements (refractory metals characterized by a long time of diffusion in matter and by a low probability of ionization).

Wide perspectives are open in the study of these nuclei by using kinematic separators on the heavy ion beams. Produced in the reactions nuclei recoils are used in these set-ups. The velocity of these recoils is high enough (as a rule more than 10^9cm/s), and the distance of several meters (between the place of their production and the spectrometer) is covered during the time of less than 10^{-7} s. There is no ion source in these set-ups, and, thus, the limitations for the pointed out above elements are removed.

Different nuclear reactions are used for the studied nuclide production. The beams of the produced nuclei have a low angular divergence and a moderate energy spread. These beams are successfully used in a number of heavy ion accelerators, for example in GANIL (France) [1], RIKEN (Japan) [2], MSU (USA) [3], CYCLONE (Belgium). With the use of these beams nuclides far from stability line and nuclear reactions are being studied. A similar set-up is built at the cyclotrons of the Flerov Laboratory of Nuclear Reactions, JINR [4,5].

It is very interesting to use the beams of radioactive nuclei for the determination of the pointed above nuclear parameters $\Delta\langle r^2\rangle$, μ and Q by laser spectroscopy methods. The intensity of the beams is sufficient for laser experiments. But these beams consist of multiple charged ions and their excited levels are too high for laser radiation. However, some peculiarities of the multiple charged ions allow one to overcome these difficulties. In a number of ions there are long-lived metastable levels and it is possible to use these levels for laser excitation.

In the present paper the perspectives are discussed for the study of nuclear structure by the laser spectroscopy methods on the beams of accelerated radioactive nuclei. The calculations of the isotope shifts and hyperfine splitting for the ions with one or two electrons are performed and the experimental set-up for the study of these ions is described.

LEVELS OF HYDROGEN-LIKE AND HELIUM-LIKE IONS. Long-lived metastable states suitable for the excitation of the other levels by laser radiation are observed in the ions with one or two electrons (hydrogen-like and helium-like ions, respectively). The level schemes including these excited states are presented in Fig.1. The calculation of the energies of these levels has been performed for H-like [6] and He-like [7] ions. As it is seen from Fig.1, the metastable levels are $2^2S_{1/2}$ in H-like and 2^1S_0, 2^3S_1 in He-like ions. The energies of these levels as a function of the element atomic number are shown in Fig.2. They are close to the KX-ray energies for the corresponding elements.

E1 - transitions between the metastable and ground states ($1^2S_{1/2}$ or 1^1S_0) are forbidden by the selection rule. Therefore, the main mode of decay is the emission of two electric dipole quanta for the $2^2S_{1/2}$ ($Z<45$) and 2^1S_0 (all Z) levels or the emission of magnetic dipole quanta for the 2^3S_1 level. Half-lives of the metastable levels have been calculated by the random phase method in the non-relativistic approximation [8] and presented in Fig.3. It is seen that these half-lives are long enough - $\tau \geq 10^{-9}$ s at $Z \leq 22$ for the $2^2S_{1/2}$ and 2^1S_0 levels or at $Z \leq 30$ for the 2^3S_1 levels. These half-lives are sufficient for the ions to fly several sentimetres before their decay.

As it can be seen from Fig.1, the excited p-levels are situited close to the metastable states ($2^2P_{1/2}$ and $2^2P_{3/2}$ in H-like ions, 2^1P_1, 2^3P_J in He-like ions). It is possible to excite these levels from the metastable states by laser radiation of the optical wave-length range. The required for this excitation wave-lengths are presented in Table 1. They are accessible for lasers of different types. It is convenient to use He-like ions for the light elements (He - Ne, $2^1S_0 \to 2^1P_1$ and $2^3S_1 \to 2^3P_0$ transitions) and H-like ions for more heavy ones (Ne - Ar, $2^2S_{1/2} \to 2^2P_{3/2}$ transition, Ca - Sn, $^2S_{1/2} \to {}^2P_{1/2}$).

Since the parity of p-levels is negative, these levels are decayed to the ground 1S and to the metastable 2S states by E1-transition. In the first case the energy of the decay is close to the energy of the metastable states and for the elements with $Z \geq 10$ it is more than 1 keV. This value is

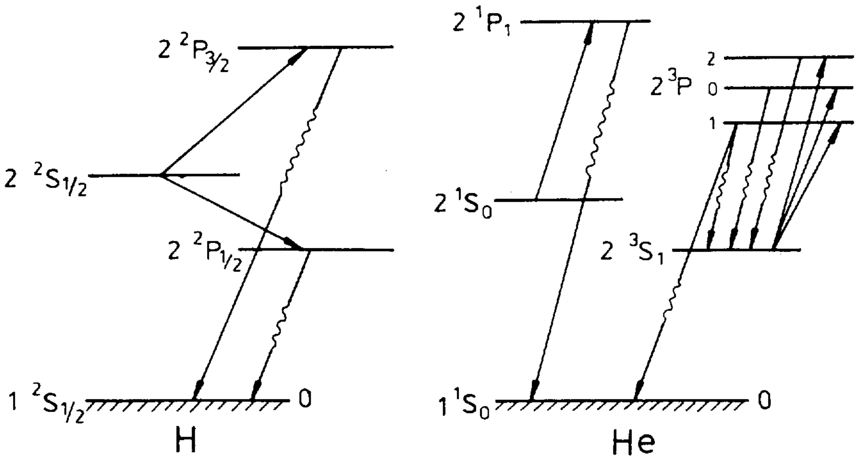

Fig. 1. Schemes of levels and radiation transitions in H-like and He-like ions.

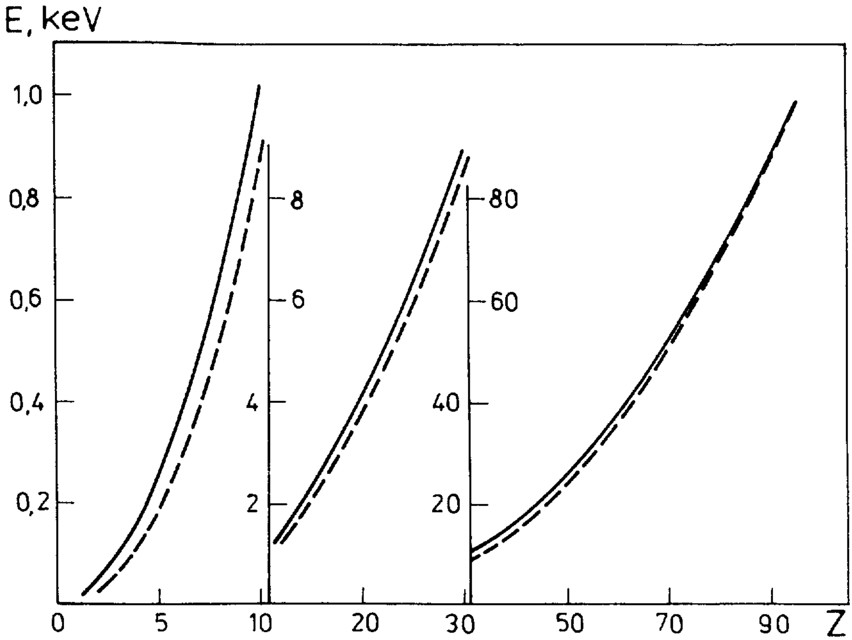

Fig. 2. Metastable levels energy in H-like (solid line) and He-like (dashed line) ions plotted against the atomic number of the element Z.

much more than the energy of laser radiation. Hence it is possible to separate both kinds of radiation and to decrease the background of the detector.

PRODUCTION OF METASTABLE H- AND HE-LIKE IONS. The equilibrium ion charge during the movement of ion in matter depends on its kinetic energy. The value of the mean charge $\bar{q}$ and its dispersion $\sigma(q)$ is determined by the relations [9]:

$$\bar{q} = Z[1-1,034\exp(-\frac{V}{V_o Z^{0.688}})], \tag{1}$$

$$\sigma(q) = 0.15\sqrt{Z}, \tag{2}$$

where Z and V - are the atomic number and the velocity of ion, $V_o = 2.19 \cdot 10^9$ cm/s is the velocity of electron at the first orbit of H-atom. It is possible to determine the kinetic energy of ions, when they become H-like (q=Z-1) or He-like (q=Z-2) ions using the relation (1). This energy as a function of the ion atomic number is presented in Fig.4. It is seen that the range of this energy varies from 0.01 MeV/nucl for He-like Li up to 250 MeV/nucl for H-like U. But in any case it can be obtained at modern heavy ion accelerators.

It is necessary to populate the metastable states in the ions for the laser excitation of p-levels. The experiments showed, that the largest probability of the metastable level population (up to 10%) was obtained at the pick-up of one or two electrons by the fully stripped ions [10].

ISOTOPE SHIFTS AND HYPERFINE SPLITTING. The calculations of the levels of H-like ion have been performed on the basis of Dirac equation for the movement of a single electron in the Coulomb field of a nucleus. For obtaining the precise values a number of corrections was included: relativistic effects, vacuum polarization, reduced mass of the electron-nucleus system, finite sizes of a nucleus. The two latter corrections are the most sensitive to the isotopic variations. They can be used for the determination of the corresponding nuclear parameters (the mass and charge radius). The reduced mass of a nucleus has been taken into consideration in the correction to the energy level:

$$\Delta E_M = \frac{m}{m + M}[1 + \frac{(Z\alpha)^2}{4n^2}]E, \tag{3}$$

where m is the mass of the electron, Z and M are the atomic number and the mass of the nucleus, E and n - the energy and the principal quantum number

TABLE 1

Wave-lengths of laser radiation for the excitation 2P-levels from the metastable states

Element	λ($S_{1/2}\rightarrow P_{3/2}$) nm	λ($S_{1/2}\rightarrow P_{1/2}$) nm	λ($^1S_0\rightarrow {}^1P_1$) nm	λ($^3S_1\rightarrow {}^3P_0$) nm
He			2058	1083
Li			956	548
Be			617	372
B			448	283
C			353	227
N			288	191
O	6990		246	164
F	4480		212	144
Ne	2850		196	128
Na	1940		165	
Mg	1365		147	
Al	990			
Si	740			
P	556			
S	450			
Ar	266	7825		
Ca	174	5430		
Zn		1345		
Br		795		
Zr		500		
Rh		332		
Sn		229		

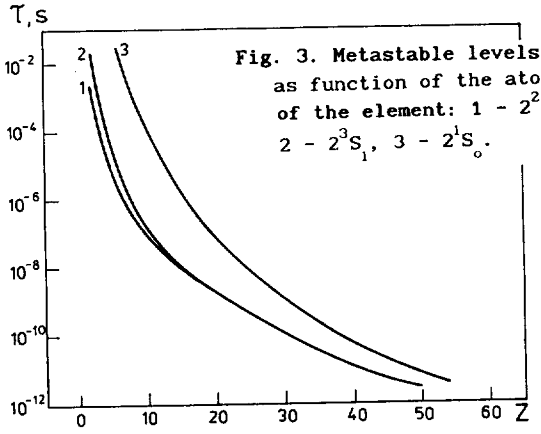

Fig. 3. Metastable levels half-life as function of the atomic number of the element: 1 - $2^2S_{1/2}$, 2 - 2^3S_1, 3 - 2^1S_0.

of the level, α - the fine structure constant. Neglecting the relativistic effects the shift of the resonance frequency for two isotopes of M_1 and M_2 masses (the normal mass shift) is:

$$\Delta\nu = \frac{c}{\lambda} \cdot \frac{m(M_1 - M_2)}{M_1 \cdot M_2}, \tag{4}$$

where λ is the wave-length of the radiation transition, c - the rate of the light.

In H-like ions with a single electron the specific mass shift induced by the correlated movement of the electrons in the Coulomb field of the nucleus and characteristic of the multielectron atoms is absent. Thus, only the normal mass shift determined by eq. (4) is important. A number of mass shift examples is presented in Table 2.

The shift of the energy level of the atom or the ion induced by spatial distribution of the electric charge of the nucleus (the field shift) depends on its principal and orbital numbers. In the case of the considered above H-like and He-like ions the shift of the 2S-levels is the most important one. The value of the field shift for the $2S_{1/2} \rightarrow 2P_{1/2,3/2}$ transition is determined by the relation:

$$\Delta E_{fs}=[1+1.38(Z\alpha)^2]\,\frac{(Z\alpha)^2}{12}\left(\frac{Z\alpha R}{\lambda_c}\right)^{2S} mc^2, \tag{5}$$

where R - is the charge radius of the nucleus, λ_c - the Compton wave-length of the electron. The field shift for the two isotopes with different mean-square charge radii ($\langle r^2\rangle_1 - \langle r^2\rangle_2 = \Delta\langle r^2\rangle$, where $\langle r^2\rangle = 3/5R^2$) is expressed as follows:

$$\Delta\nu_{fs} = 1,2\cdot 10^{-4} Z^4 \Delta\langle r^2\rangle \ \mathrm{GHZ/fm^2}, \tag{6}$$

The calculated dependence of the field shift at $\Delta\langle r^2\rangle = 1\ \mathrm{fm}^2$ on the atomic number Z of ions is shown in Fig.5, and a number of examples - in Table 2. The field shifts of the neutral atoms of the alkali elements (Li, Na, K, Rb, Cs) at the same $\Delta\langle r^2\rangle$ are shown for the sake of comparision. It is seen that the ion field shift is much larger than that of the neutral atom, and the difference grows rapidly with the increasing of Z. This phenomenon is evidently explained by a short distance between the 2S-electrons and the nucleus and a more strong interaction between them.

He-like ions with two electrons require more complicated calculations. Two steps of the calculations have been performed. Firstly, the non-relativistic wave-function was obtained by the Hartree-Fock method for 1S, 2S and 2P electrons. Secondly, the Dirac equation was solved for these

TABLE 2

Isotopic shifts and Doppler broadening in H-like and He-like ions

Element	Ion	Transition	$\Delta\nu_{FS}$ GHz/fm^2	$\Delta\nu_{MS}$ GHz	$\Delta\nu_{D1}$ GHz	$\Delta\nu_{D2}$ GHz
Li	He-	${}^3S_1 \to {}^3P_0$	0,0047	24	45	0,50
Be	He-	${}^3S_1 \to {}^3P_0$	0,020	32	120	1,8
B	He-	${}^3S_1 \to {}^3P_0$	0,062	39	220	4,8
C	He-	${}^3S_1 \to {}^3P_0$	0,154	50	360	10
O	He-	${}^1S_0 \to {}^1P_1$	0,803	54	525	22
Ne	He-	${}^1S_0 \to {}^1P_1$	1,44	59	1230	98
	H-	$S_{1/2} \to P_{3/2}$	1,98	0,14	84	6,7
Mg	H-	$S_{1/2} \to P_{3/2}$	4,01	0,20	200	17
Si	H-	$S_{1/2} \to P_{3/2}$	7,37	0,27	460	50
S	H-	$S_{1/2} \to P_{3/2}$	12,2	0,36	870	110
Ar	H-	$S_{1/2} \to P_{3/2}$	21,8	0,49	1700	255
Zn	H-	$S_{1/2} \to P_{1/2}$	130	0,029	550	140
Br	H-	$S_{1/2} \to P_{1/2}$	290	0,032	1080	320
Zr	H-	$S_{1/2} \to P_{1/2}$	500	0,035	1900	600
Rh	H-	$S_{1/2} \to P_{1/2}$	990	0,042	3250	1350

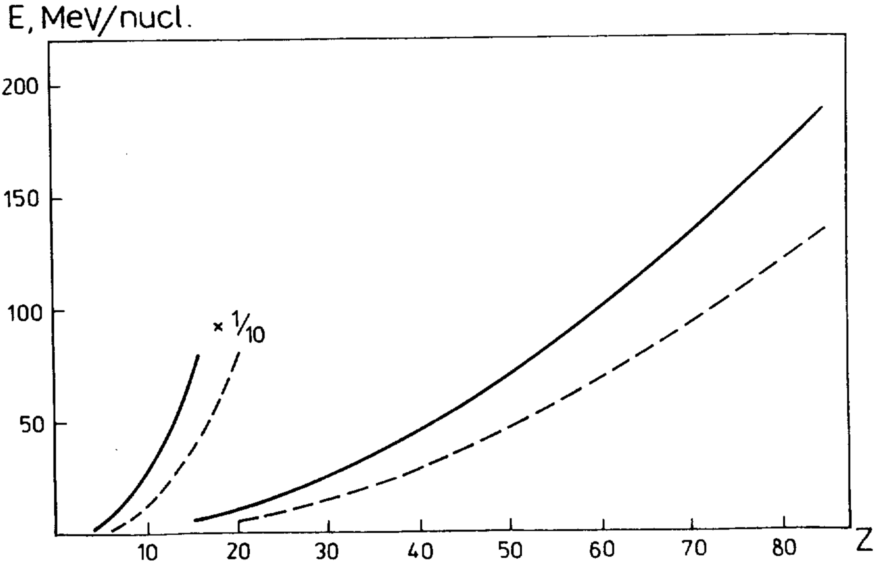

Fig. 4. Dependence of the energy required for production of H-like (solid line) and He-like (dashed line) ions on the atomic number of the element.

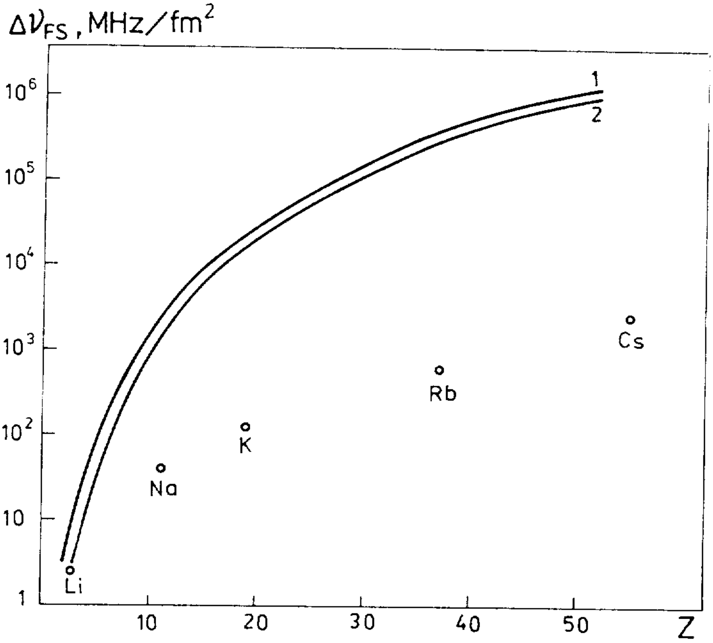

Fig. 5. Dependence of the field shift on the atomic number of the element; 1 - H-like ions, 2 - He-like ions, dots - neutral atoms.

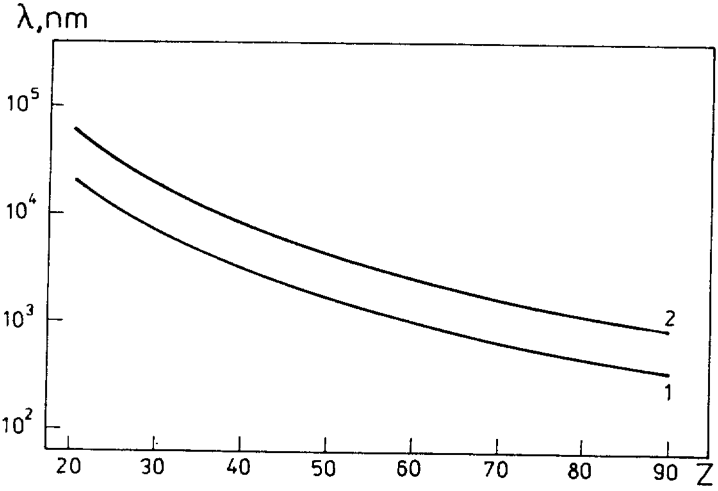

Fig. 6. Wave-length of the radiation transition between the component of $^1S_{1/2}$ state in H-like ion against the atomic number of the element for odd-proton - 1 and odd-neutron -2 nuclei.

electron configurations in the self-consistent Coulomb field. The field shift and the specific mass shift were calculated this way. The field shift dependence on Z is shown in Fig.5 and in Table 2. It is seen that there is a small difference in the field shifts of He-like and H-like ions.

The examples of He-like ion mass shifts are presented in Table 2 at $\Delta A=1$ (the sum of normal and specific mass shifts). The mass shifts of H-like and He-like ions are large and it is necessary to use precise values of the nuclei masses for the mass correction.

A small distance between the nucleus and 1S-electron in H-like ions induces a strong hyperfine splitting. The value of this splitting (the difference between the component energies with the total moments $F_1=I+1/2$ and $F_2=I-1/2$, where I is the spin of the nucleus) is [11]:

$$\Delta E(F_1,F_2)=\frac{4}{3}\,\frac{(2I+1)hcR_Y\alpha^2Z^3\mu}{I}, \tag{7}$$

where R_Y is Rydberg constant, μ - magnetic dipole moment of the nucleus. The wave-lengths of the radiation transitions between the 1S-level component of the hyperfine splitting in H-like ions are presented in Fig.6. The following values of magnetic moments from the Schmidt model [12] for the Z-odd and N-odd nuclei and I=5/2 are used:

$$\begin{aligned}\mu_p&=4,79\mu_B\\ \mu_n&=-1,91\mu_B,\end{aligned} \tag{8}$$

where μ_B is the nuclear magneton. A fast decreasing of the wave-length values with increasing Z is observed. For the elements with Z>50 the wave-length is found in the range of optical radiation, and the excitation by laser is possible. Precise measurements of these wave-lengths allow one to determine the values of magnetic moments with a high accuracy and to obtain the information about the spatial distribution of the electric current in the nucleus.

DOPPLER SHIFTS OF THE OPTICAL LINES. As it is known, resonance frequency of the moving atom or ion is shifted (Doppler effect). The value of this shift in the first approximation (the linear Doppler effect) is:

$$\Delta\nu=\nu\frac{v}{c}\cos\alpha, \tag{9}$$

where ν is the resonance frequency, v - the ion velocity, α - the angle between the directions of the ion and laser ray. The linear Doppler shift is minimal if both directions are mutually orthogonal ($\alpha=90^\circ$). Only the broadening of the resonance frequency induced by the angular divergency of

TABLE 3

Radiative widths of 2P-livels in H-like and He-like ions

Element	Γ(2P), GHz				
	$P_{3/2}\to 1S_{1/2}$	$^1P_1\to 1^1S_0$	$^3P_0\to 2^3S_1$	$^3P_1\to 1^1S_0$ 2^3S_1	$^3P_2\to 1^1S_0$ 2^3S_1
Li	$6,1\cdot 10^{1}$	$1,4\cdot 10^{1}$	$2,5\cdot 10^{-2}$	$2,5\cdot 10^{-2}$	$2,5\cdot 10^{-2}$
Be	$1,6\cdot 10^{2}$	$1,2\cdot 10^{2}$	$3,6\cdot 10^{-2}$	$3,6\cdot 10^{-2}$	$3,6\cdot 10^{-2}$
C	$8,1\cdot 10^{2}$	$8,8\cdot 10^{2}$	$5,8\cdot 10^{-2}$	$8,7\cdot 10^{-2}$	$5,8\cdot 10^{-2}$
O	$2,6\cdot 10^{3}$	$3,3\cdot 10^{3}$	$8,0\cdot 10^{-2}$	$6,3\cdot 10^{-1}$	$8,3\cdot 10^{-2}$
Ne	$6,2\cdot 10^{3}$	$8,8\cdot 10^{3}$	$1,0\cdot 10^{-1}$	$5,5\cdot 10^{0}$	$1,1\cdot 10^{-1}$
Mg	$1,3\cdot 10^{4}$	$1,9\cdot 10^{4}$	$1,4\cdot 10^{-1}$	$3,4\cdot 10^{1}$	$1,8\cdot 10^{-1}$
Si	$2,4\cdot 10^{4}$	$3,8\cdot 10^{4}$	$1,7\cdot 10^{-1}$	$1,6\cdot 10^{2}$	$2,3\cdot 10^{-1}$
S	$4,1\cdot 10^{4}$	$6,2\cdot 10^{4}$	$2,0\cdot 10^{-1}$	$5,8\cdot 10^{2}$	$3,9\cdot 10^{-1}$
Ar	$6,6\cdot 10^{4}$	$1,1\cdot 10^{5}$	$2,3\cdot 10^{-1}$	$1,8\cdot 10^{3}$	$6,6\cdot 10^{-1}$
Ca	$1,0\cdot 10^{5}$	$1,5\cdot 10^{5}$	$2,6\cdot 10^{-1}$	$4,8\cdot 10^{3}$	$1,2\cdot 10^{0}$
Zn	$5,1\cdot 10^{5}$	$7,8\cdot 10^{5}$	$5,0\cdot 10^{-1}$	$1,2\cdot 10^{5}$	$2,4\cdot 10^{1}$
Zr	$1,6\cdot 10^{6}$	$2,4\cdot 10^{6}$	$9,1\cdot 10^{-1}$	$5,8\cdot 10^{5}$	$2,0\cdot 10^{2}$
Sn	$3,9\cdot 10^{6}$	$5,0\cdot 10^{6}$	$1,7\cdot 10^{0}$	$2,1\cdot 10^{6}$	$1,4\cdot 10^{3}$

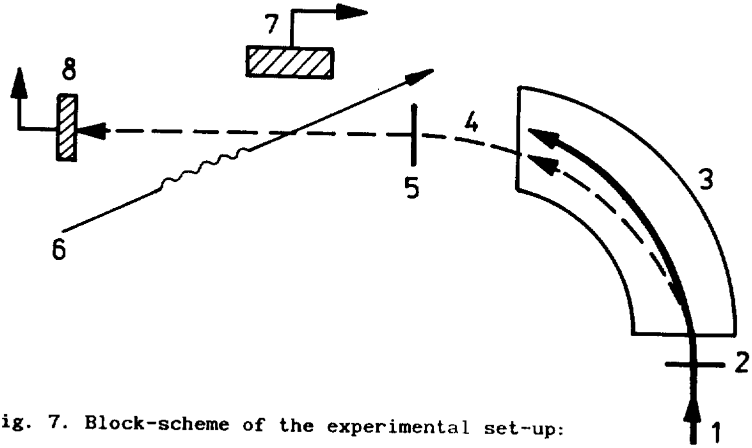

Fig. 7. Block-scheme of the experimental set-up: 1 - beam of accelerated ions, 2 - target, 3 - kinematic separator, 4 - beam of studied isotopes, 5 - carbon film, 6 - laser ray, 7 - detector of resonance fluorescence radiation, 8 - position-sensitive detector of ions.

the ion beam ($\Delta\alpha$) takes place:

$$\delta(\Delta\nu_{D1})=\nu\frac{v}{c}\Delta\alpha. \qquad (10)$$

The calculated Doppler broadenings at the velocities required for production of H-like and He-like ions and at $\Delta\alpha=1\%$ are presented in Table 3. As it can be seen, these broadenings are large enough, they are in many cases larger than the mass and field shifts and the widths of the excited p-levels. That is why it is necessary to use the ion beams with a minimal angular divergency ($\Delta\alpha<1\%$).

However, the high velocity of the ions induces the second order effect (quadratic Doppler effect):

$$\Delta\nu_{D2}=\nu\frac{v^2}{c^2}. \qquad (11)$$

Then in the parallel ion beam (cos $\alpha=0$) the resonance frequency broadening will be induced by the energy dispersion of the beam $\Delta E/E$:

$$\delta(\Delta\nu_{D2})=\nu\frac{v^2}{c^2}\frac{\Delta E}{E}. \qquad (12)$$

The resonance frequency broadenings induced by the quadratic Doppler effect at $\Delta E/E=1\%$ are presented in Table 3. It is seen that these broadenings are less and become noticeable only in the most heavy ions.

CROSS-SECTIONS OF P-LEVELS EXCITATION. The cross-section for the resonance excitation of atomic or ionic levels is determined by the Breit-Wigner formula [13]:

$$\sigma = \frac{\lambda^2}{4\pi}\cdot\frac{(2I_f+1)}{(2I_i+1)}\cdot\frac{\Gamma_f\Gamma_o}{(E-E_o)+\Gamma_o^2}, \qquad (13)$$

where λ is the wave-length of laser radiation, I_i and I_f - the spins of the initial and final states, Γ_f and Γ_o - the partial and total widths of the excited level, E_o and E - the energy of the level and laser radiation. In the case of resonance ($E=E_o$) σ is equal to:

$$\sigma = \frac{\lambda^2}{4\pi}\cdot\frac{(2I_f+1)}{(2I_i+1)}\cdot\frac{\Gamma_f}{\Gamma_o}. \qquad (14)$$

It is seen that the cross-section strongly depends on the ratio of partial and total widths. The values of the partial widths for p-levels in H-like and He-like ions were calculated by the random phase method.

The total width consists of the homogeneous and inhomogeneous widts. The first one includes all possible modes of decay. The most important one is the decay to the $1S_{1/2}$ or $1S_o$ ground state. The $^2P_{1/2}$ and $^2P_{3/2}$ levels in H-like ions and $^1P_1$ in He-like ions undergo this mode decay (E1 -

transition). The half-life for these transitions is determined by the relation [15]:

$$\tau(E1)=1{,}6\cdot10^{-9}Z^{-4}. \qquad (15)$$

The corresponding to this half-life total widths are presented in Table 3. They are much longer than the partial widths for the decay to metastable state (the ratios $\Gamma_f/\Gamma_o \simeq 10^{-6}-10^{-4}$). In these cases the cross-sections for the excitation p-levels are occured in the range of $10^{-14}-10^{-16}$ cm^2.

The E1 transitions from the $^3P_0$ and $^3P_2$ levels to the 1^1S_0 ground state are forbidden and from the $^3P_1$ level are delayed. In the cases of these levels the widths for the transitions to the ground state are small and therefore the ratio is $\Gamma_f/\Gamma_o \simeq 1$. But the contribution of inhomogeneous widths becomes important. These widths are determined mostly by the Doppler broadening (Table 3). The cross-section of the excitation of these levels depends on the ion beam parameters (its angular and energy resolution).

EXPERIMENTAL SET-UP. The scheme of the experimental set-up for the measurements of resonance frequences on the beams of accelerated H-like and He-like ions is presented in Fig.7. The operation of this set-up is based on the excitation of the levels by the optical laser radiation and on the detection of the spontaneous X-rays.

The studied isotopes are produced in the interaction of the accelerated in the cyclotron ions with the nuclei of the target. The most effective method of isotope production is the bombarding ion fragmentation, in which the reaction products move in the direction of the beam. The selection of the studied isotopes is performed by the kinematic separator, consisting of a number of magnetic dipoles and quadrupoles and a degrader [4,5]. Two or three neighbouring isotopes of the same element are selected by the kinematic separator for the resonance frequency measurement. The kinetic energy of these isotopes is chosen to obtain the H-like or He-like ions after their passage through a thin ($\simeq 20\mu g/cm^2$) carbon film, positioned 1 cm before the laser ray.

Part of the ions after the passage through the film transit to the metastable ($2S_{1/2}$, $2S_0$ or $2S_1$) state. It is possible to excite these metastable ions by laser radiation. But the angular and energy dispersion of the studied isotope beam induces a broadening of the resonance line of up to some thousand GHz. It is difficult to measure the shape of such a broad line with a requisite accuracy.

It is possible to overcome these difficulties if one uses another method for the resonance frequences measurement. The laser frequency is not scanned but fixed and the energy and angle of the excited ions are measured [16]. These measurements allow one to determine the Doppler shifts of the frequencies,using expressions (9) and (11), and to obtain the values of the resonance frequencies for the studied isotopes.

This method is used in the presented experimental set-up. A position sensitive silicon detector is used for the measurements of the energy and the angle for the excited ions and a photomultiplier or a gaseous X=ray detector - for the resonance scattered radiation. The spectrum of the coincidences between the ions and scattered radiation is measured.

CONCLUSION. Measurements of the isotopic shifts and hyperfine splitting in the optical spectra of H-like and He-like ions open new perspectives in the study of nuclear structure. The using of such accelerated ions allows one to extend these investigations to the earlier inaccessible regions of elements, for example, groups V-VII of the Mendeleev's periodic table. In the atoms of these elements the sensitive to the electric charge distribution levels are situated too high to be excited by laser radiation Moreover, complicated electronic configurations of these levels do not allow one to obtain the correct values of the charge radii from the measured isotopic shifts.

In contrast, H-like and He-like ions have a simple and the same for all elements system of levels. And it is easy to extract the nuclear parameters from the experimental data. The proposed experimental set-up is suitable for the study of the most short-lived nuclei. Therefore, the method based on the using of H-like and He-like accelerated ions in the secondary nuclei beam is a perspective one, despite the complicated experimental technique.

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The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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