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Laser Method of Highly Selective Detection of Rare Radioactive Isotopes through Multistep Photoionization of Accelerated Atoms

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Abstract. A laser detection method is proposed for ultralow abundance ($<10^{-10}$) of rare radioactive isotopes based on a multistep photoionization of accelerated atoms with a selectivity up to 10^7 on each step of a collinear laser excitation.

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In solving many problems of nuclear physics, geophysics, cosmochronology, etc. it is necessary to determine ultralow concentrations of long-lived radioactive isotopes with a very high selectivity (to 10^{22}) in the presence of the stable isotopes of the same element. Much the same problems arise in: 1) detecting isotopes of cosmogenic origin, particularly beryllium-10 (with its half-life $T_{1/2}=2.7\times10^6$ years), aluminium-26 ($T_{1/2}=7.4\times10^5$ years), chlorine-36 ($T_{1/2}=3.1\times10^5$ years) and so on, their natural relative abundance being, respectively, 10^{-10}, 10^{-14}, and 10^{-17}; 2) developing a solar neutrino detector when, for example, the ^{41}Ca isotope must be detected with selectivity of 10^{21} [1]. The use of the methods of nuclear physics techniques based on the observation of the radioactive decay by low level counting to detect such rare isotopes is inefficient because of high values of $T_{1/2}$.

Recently the direct detection of such a rare isotope (as ^{14}C, ^{10}Be, ^{36}Cl) with the use of Van de Graaf accelerators [2, 3] and cyclotrons [4] has been demonstrated.

The methods of laser detection of single atoms [5–7] and isotopes based on selective photoionization or fluorescence are under intense development now.

The maximum selectivity of laser excitation of a rare isotope against the background of the basic isotope is determined by the inevitable absorption at an absorption line wing of the basic isotope and equals [5, 8]:

$$S_0=\left(\frac{\Delta\nu_{is}}{\Gamma}\right)^2, \tag{1}$$

where $\Delta\nu_{is}$ is the isotopic shift, Γ is the homogeneous natural line width (HWHM). Such very slight overlapping of the spectral line wings makes it impossible to obtain very high critical values of selective detection of the above-mentioned cosmogenic isotopes. The situation is more complicated for elements with a small isotope shift that can be comparable to the Doppler broadening.

Among the methods of laser spectroscopy there have been developed very efficient techniques for narrowing Doppler-broadened spectral lines. This pool has been lately extended by an effective method of collinear laser spectroscopy of accelerated atoms [9]. An important advantage of this method consists of the velocity-bunching phenomenon of ions during their acceleration. In this linear method of laser spectroscopy the Doppler line width can be reduced to the natural one.

In this letter paper particular emphasis is placed, first, on the occurrence of an isotope "mass" Doppler shift for all spectral lines of an accelerated atom and, secondly, on the resulting possibility of highly selective multistep excitation and ionization of accelerated atoms. The occurence of an additional isotopic shift of any spectral line can help to realize the previously proposed idea of selectivity multiplication at multistep excitation and ionization [8]. Thus, a combination of the methods of collinear laser spectroscopy and multistep photoionization proves to be very useful. We are going to consider this possibility in more detail.

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The Proposed Method

According to the method of collinear laser spectroscopy [9], the atoms with thermal velocities are first converted to ions, then the ions are accelerated by the electric field and, at last, they are neutralized back.

After passing the potential difference U the ions with their masses M_1 and M_2 have different velocities $\Delta v_{M_1M_2}=\sqrt{2eU}\,(1/\sqrt{M_1}-1/\sqrt{M_2})$. Then the accelerated ions are neutralized in the charge-exchange cell. Since the charge-transfer cross-sections are much larger than the collisional cross-section the neutralized beam will not scatter and its velocity will be almost equal to that of the ion beam [10]. In this case the shift due to different velocities of the atoms M_1 and M_2 will be

$$\Delta\nu^v=(\nu_0^{M_1}-\nu_0^{M_2})\pm\frac{v_1\nu_0^{M_1}-v_2\nu_0^{M_2}}{c}, \qquad (2)$$

where $\nu_0^{M_1}$ and $\nu_0^{M_2}$ are the nonshifted transition frequencies of the isotopes M_1 and M_2, c is the light velocity. Assuming that $\nu_0^{M_1}=\nu_0^{M_2}$ we obtain expression for the value of additional isotope shift

$$\Delta\nu^v=\pm\frac{\nu_0}{c}\sqrt{2eU}\left(\frac{1}{\sqrt{M_1}}-\frac{1}{\sqrt{M_2}}\right). \qquad (3)$$

The plus or minus signs correspond to the different directions of propagation of the laser radiation and the atomic beam. Figure 1 shows the dependences of the additional isotope shift $\Delta\nu^v$ at varied accelerating voltage U and ordinary isotope shift ($\Delta\nu_{is}/\nu\sim 1/M^2$) on atomic mass for the case $M_2-M_1=1$. It may be seen that even at $U=10$ keV the artificial isotopic shift is twenty times larger than the ordinary one. By choosing a proper direction of the atomic beam and the laser radiation we can use both isotope shifts. Such a method of excitations is especially important when the atomic transition in use has not an ordinary isotope shift.

The acceleration of the atomic beam causes the Doppler line to be narrowed by a factor of 1/2 $(kT/eU)^{1/2}$ [9]. When the ion source temperature $T=1500$ K and the accelerating voltage is 10 kV this value is 1.8×10^{-3}. The Doppler width the resonant aluminium line ($\lambda=3944$ Å) at $T=1500$ K equals 4.1 GHz, and after being accelerated it is narrowed to 7.4 MHz that is less than the radiative line width $2\Gamma=7.8$ MHz. Thus, when collinearly excited the accelerated atoms not only eliminate the Doppler broadening [9] but also give rise to an additional isotope shift in the absorption spectra.

For illustration, let us consider the possibility of detection of aluminium-26 through multistep ionization. Natural aluminium has one stable isotope ^{27}Al.

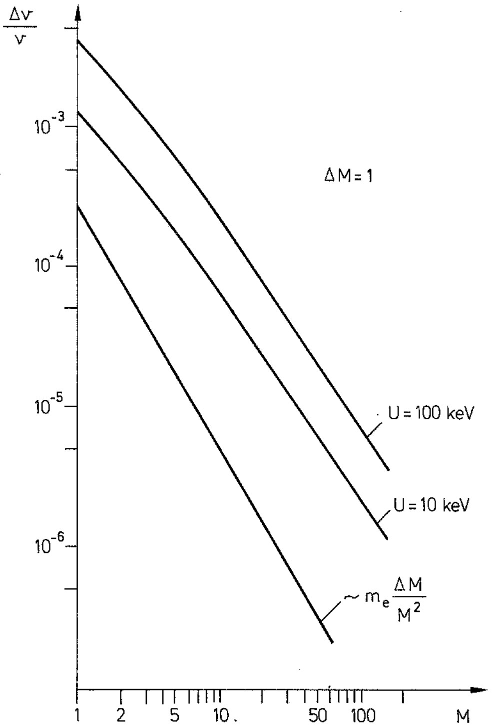

Fig. 1. The ordinary ($\sim 1/M^2$) and additional isotope shifts for atoms with energies of 10 keV and 100 keV as a function of atomic mass

The steadystate abundance of radioactive ^{26}Al is 10^{-14}. So the ionization selectivity must be much higher than 10^{14} to detect ^{26}Al. Figure 2a schematically shows the levels of the aluminium atom and the transitions which can be used. The transition wavelengths lie within a spectral range accessible for dye lasers. Ionization is performed through the autoionization state $3s3p^2\,{}^3P$ [11]. Figure 2b shows how the absorption spectrum at the $3p-4s$ transition differs for the thermal $T=1500$ K (left) and accelerated up to 10 keV atoms (right). For the thermal atoms the isotope shift (590 MHz) is smaller than the hyperfine splitting (750 MHz) and much smaller than the Doppler line width (4.1 GHz). For the accelerated atoms the additional isotope shift equals 12 GHz, and the Doppler line width becomes smaller than the radiative width. Let us estimate the selectivity of excitation at the first step assuming that the excitation is performed by laser radiation with a spectrum width $\Delta\nu_l$ smaller than 2Γ. In this case the selectivity S_1 is determined by (1) and equals 9.5×10^6. The excitation selectivity at the second step $S_2\simeq S_1$. The transition line width during excitation to an autoionization state is 230 GHz [11]. That is much larger than the isotopic shift and hence $S_3=1$. The total ionization selectivity is equal to the product of selectivities at each step $S=S_1S_2\cdot S_3=9\times10^{13}$. By carrying out separation also by a mass-spectrometer with selectivity $S_{MS}=10^2$ we can obtain

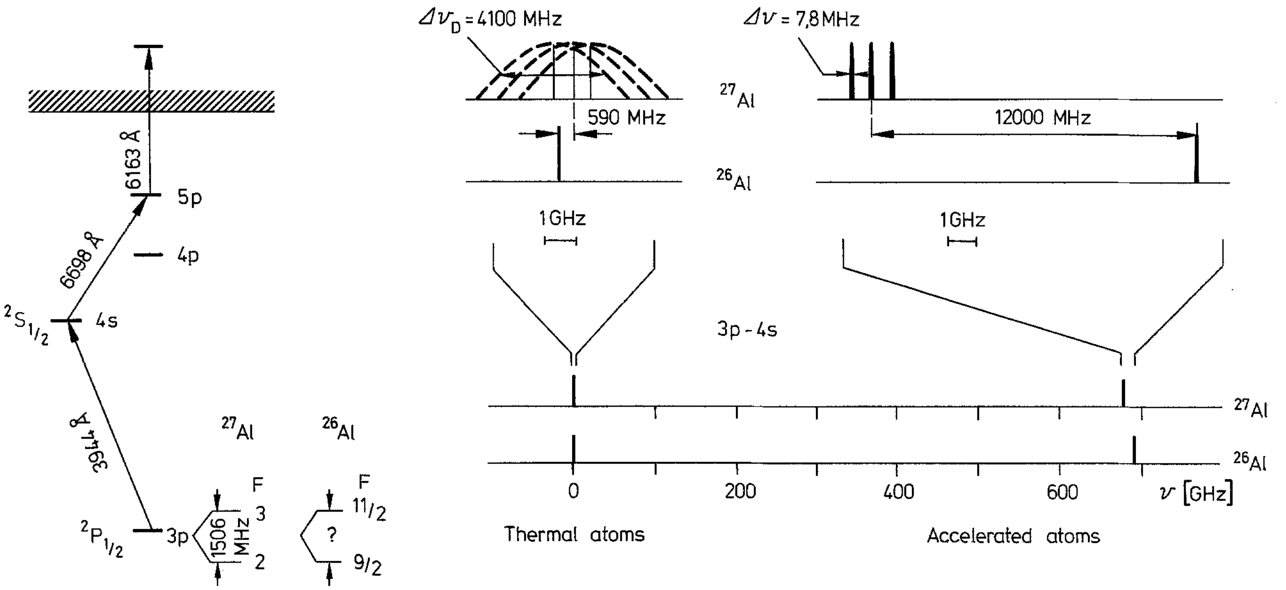

Fig. 2. (a) The scheme of aluminium atomic levels and the transition wavelengths which can be used to detect the ^{26}Al isotope. (b) The absorption line structure of the ^{26}Al and ^{27}Al isotopes at the $3p$–$4s$ transition. Left: ordinary observation; right: collinear observation of atoms accelerated to 10 keV. The hyperfine structure constants are known only for ^{27}Al($I = 5/2$), Al($^2S_{1/2}$) = Al($^2P_{1/2}$) = 502 MHz [12]. The absorption of ^{26}Al($I = 5$) shown is hypothetical

the resulting recording selectivity of ^{26}Al against ^{27}Al 9×10^{15}.

It is possible to obtain selectivity at the ionization step with a collinear excitation if the atom passes to a long-lived autoionization or Rydberg state. But there is no selectivity when it is ionized to a continuum or to an autoionization state with the spectral width larger than the isotope shift. In this case, taking into account the fact that ions with different mass in a pulsed operational mode on the step of ionization are separated in space, we can direct the radiation at the third frequency ν_3 at right angle to the beam axis at a proper instant of time and obtain additional selectivity.

The change of the Al atom energy by 1 eV will correspond to the change of resonant frequency by 34 MHz. Therefore, to realize the maximum excitation selectivity, one should use a monoenergetic atomic beam. In case of atoms with their ionization potential being below 6 eV a surface-ionization source can be used [13]. The ion energy spread at the source temperature 2300 K will come to 0.2 eV. Another cause of energy distribution broadening is a nonresonant charge-exchange of the ion beam neutralized. The broadening value in this case may be as high as several electron-volts [9].

To produce aluminium ions (ionization potential: 5.99 eV) one can use surface ionization on plantinum ($\varphi = 5.3$ eV). Charge-exchange of aluminium ions can be done in lithium vapour (ionization potential: 5.39 eV), the energy defect being 0.6 eV.

The main problem in recording such high selectivity consists of producing a large ion flow. With an ion current of $I = 10^{-4}$ A about 6×10^{17} ions can be passed in a measurement time of 10^3 s with 6×10^3 ions of ^{26}Al.

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