Optical excitation of isotopes that are selective in nuclear spin

Ya. B. Zel'dovich and I. I. Sobel'man

P. N. Lebedev Physics Institute, USSR Academy of Sciences
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We consider the possibility of atomic excitation that is selective in nuclear spin; this possibility is connected with lifting the hindrance to radiative transitions as a result of hyperfine interaction.

Laser methods of isotope separation, as is well known, are based on the possibility of selective excitation and ionization of the isotopes of some element, and also of the excitation and dissociation of the molecules, and the selectivity is attained because of the small isotopic frequency shift $\delta\omega$ of the employed transitions. [1]

In those cases when the considered isotopes have different nuclear spins, the selectivity of the photoexcitation can be ensured also by lifting the hindrance to radiative transitions, as a result of interaction of the nuclear moments.

The simplest example are transitions between levels for which the total angular momentum of the electron shell is $J = 0$. If the nuclear spin is $I = 0$, then such a transition is absolutely forbidden by the selection rule $J + J' \geq 1$. For isotopes with nuclear spin $I \neq 0$ this strong forbiddenness is lifted. The very fact that the forbiddenness is lifted as a result of a "hyperfine" interaction was discussed in the literature many times. The possibility of using this effect for isotope separation has to our knowledge never been considered before.

Let us proceed to concrete examples. We consider atoms with ground electron configuration ns^2, i.e., atoms of alkali-earth metals, and also Zn, Cd, and Hg. For isotopes of these elements with $I = 0$, the radiative transition $ns^2\ {}^1S_0 - ns\,np\ {}^3P_0$ is impossible. For isotopes with spin $I \neq 0$ there is a nonzero transition probability, since a perturbation V mixes a state ${}^1P_1$ into the state ${}^3P_0$. [1] Transitions of this type were observed earlier, e.g., for mercury Hg^{199} and Hg^{201}, λ 2655.8 Å, in[2] and for Cd, λ 3320 Å, in[3]. In the estimates of the values of the admixture χ of the state ${}^1P_1$ to the state ${}^3P_0$ it can be assumed that the main contribution to the perturbation V is made by the interaction of the s electron with the nucleus. Then

$$|\chi| = |<{}^3P_0 I | V | {}^1P_1 I> \Delta E^{-1}| = a_s \frac{\sqrt{I(I+1)}}{2\sqrt{3}\,|\Delta E|}, \quad (1)$$

where ΔE is the difference between the energies of the states ${}^3P_0$ and ${}^1P_1$, and a_s is the hyperfine-structure constant, which can be determined either from the known Fermi-Segre formulas (see, e.g., [4]) or from the experimental data on hyperfine splitting. The cross section for absorption in the ${}^1S_0 I - {}^3P_0 I$ transition is

$$\sigma = 2\pi^2 \frac{e^2}{mc}\ \frac{\omega}{\omega'}\ \frac{f}{\Delta\omega_D}\ \chi^2 = \frac{\pi^2}{6}\ \frac{e^2}{mc}\ \frac{\omega}{\omega'}\ \frac{f}{\Delta\omega_D}\ \frac{a_s I(I+1)}{\Delta E^2}, \quad (1')$$

where ω and ω' are the frequencies of the transitions ${}^1S_0 - {}^3P_0$ and ${}^1S_0 - {}^1P_1$, respectively, f is the oscillator strength of the ${}^1S_0 - {}^1P_1$ transition, and $\Delta\omega_D$ is the Doppler line width.

For the excitation to be selective it is necessary that the cross section (2) greatly exceed the cross section for absorption at the same frequency in the wing of the closest strong line ${}^1S_0 - {}^3P_1$. Denoting the ratio of these cross sections by β, we stipulate satisfaction of the inequality

$$\beta = \chi^2 \frac{f({}^1S_0 - {}^1P_1)}{f({}^1S_0 - {}^3P_1)}\ \frac{\Delta\omega^2}{\gamma\Delta\omega_D}\ 2\pi\sqrt{\pi} >> 1, \quad (2)$$

where $\Delta\omega$ is the difference between the frequencies of the transitions ${}^1S_0 - {}^3P_0$ and ${}^1S_0 - {}^3P_1$, and γ is the collision (or radiative) line width for the ${}^1S_0 - {}^3P_1$ transition.

For the mercury isotopes Hg^{199} and Hg^{201} we have $a_s \approx 1$ cm^{-1}, $\Delta E = 16\,423$ cm^{-1}, $\Delta\omega_D = 6 \times 10^9$, $f({}^1S_0 - {}^1P_1) \approx 1.25$ (see, e.g., [5]), $f({}^1S_0 - {}^3P_1) \approx 2 \times 10^{-2} f({}^1S_0 - {}^1P_1)$, [4] $E({}^3P_1) - E({}^3P_0) = 1767$ cm^{-1}, so that at $\gamma \lesssim \Delta\omega_D$ we have $\chi \approx 10^{-4}$, $\sigma \approx 10^{-19}$ cm^2, and $\beta > 10^2$. These estimates show that the photoexcitation of the ${}^3P_0$ level can be used for a rather effective separation of the isotopes Hg^{199} and Hg^{201}, with $I \neq 0$, from the even-even isotopes for which $I = 0$. [2]

Analogous estimates for other atoms with ground electron configuration ns^2 show that the described method of selective excitation of the triplet states ${}^3P_0$ can be realized also for the atoms Sr, Ba, Zn, and Cd.

Transitions $J \to J'$ with $J = J' = 0$ are not the only ones for which a "hyperfine" interaction V greatly alters the selection rules. For example, for atoms with ground configuration np^2, such as Sn and Pb, the transition from the ground level ${}^3P_0$ to one of the nearest excited levels $ns\,np^3\ {}^5S_2$ is forbidden by the selection rules for the electric dipole ($n_0 J$), and also (parity-forbidden) for the magnetic dipole and electric quadrupole. The perturbation V admixes to the ${}^5S_2$ state a ${}^3S_1$ state of the same configuration, and the J-forbiddenness for the electric dipole is lifted.

The method of selective excitation based on lifting the forbiddenness by a "hyperfine" interaction can offer in a number of cases definite advantages over methods based on the isotopic frequency shift $\delta\omega$, since it makes it possible to use pump sources that are less monochromatic and less powerful. In addition, in this case the probability of excitation transfer from isotope to another via resonant collisions is greatly suppressed, since the long-range multiple interaction are weakened by a factor χ^2 and the corresponding cross sections cannot greatly exceed πa_0^2 (we recall the the excitation—transfer cross section for an allowed dipole-dipole transition is of the order of $\pi a_0^2 (e^2/\hbar v)$, where v is the velocity of the atoms and $e^2/\hbar v \sim 10^4$). Selectivity of the excitation can therefore be assured in a wide range of pressures.

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