Role of inner electron shells of the atom in the chemical change of the probability of the converted $M4$ transition in ^{119}Sn

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To explain why stannic oxide SnO_2, unlike the metallic β-Sn, shows no measurable chemical change of the probability λ of the isomeric $M4$ transition in the ^{119}Sn nucleus ($E_\gamma = 65.3$ keV, $T_{1/2} = 293$ days), despite the large difference between the electron structure of the tin atoms in these compounds, the changes of the internal conversion coefficients (ICC) of this transition were theoretically investigated for various tin ions. The sought-for reason is the large changes of the ICC for electrons of the deep inner shells of the atoms (especially K and L). For the transition considered, the relative changes $\Delta\alpha_{nlj}/\alpha_{nlj}$ of the ICC α_{nlj} for electrons of deep inner subshells greatly exceed the relative changes $\Delta\rho_{nlj}/\rho_{nlj}$ of the electron densities ρ_{nlj} near the nucleus.

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1. The probability of the internal conversion (IC) of γ rays depends somewhat on the state of the electron shell of the atom. This leads to chemical changes of the probabilities of electromagnetic transitions in atomic nuclei. In some cases these changes can be of measurable size. An attempt was made in Ref. 1 to observe the chemical change of the decay probability of the nuclear isomer ^{119m}Sn ($T_{1/2} = 293$ days[2]), which accompanies a strongly converted transition of multipolarity $M4$ with energy 65.3 keV.[3] Unlike in the metal β-Sn, however, no measurable change was observed for the ^{119m}Sn nuclei in SnO_2, even though a considerable change of the decay probability was expected for just these compounds as a result of the large difference between the electronic structure of the tin atom in the dioxide and in the metal. The measured relative change $\Delta\lambda/\lambda$ of the decay probability λ was $+(0.1 \pm 1.2)\cdot 10^{-4}$, whereas on the basis on the data on the electronic structure of the tin atoms in SnO_2 and in β-Sn (Ref. 4) the expected change was -5×10^{-4}. This theoretical value was calculated using the customary assumption that for each electron subshell the relative change $\Delta\alpha_{nlj}/\alpha_{nlj}$ of the internal conversion coefficient (ICC) α_{nlj} is equal to the relative change $\Delta\rho_{nlj}/\rho_{nlj}$ of the electron density ρ_{nlj} near the nucleus [n is the principal quantum number, l and j are the quantum numbers of the orbital and total angular momenta, and $\rho_{nlj} \equiv \rho_{nlj}(r \to 0)$]. The large deviation of the measured near-zero $\Delta\lambda/\lambda$ from the theoretical (more than four mean squared errors) stimulated more exact calculations of the ICC for ions. They have shown that in the case of this transition the change $\Delta\alpha_{nlj}/\alpha_{nlj}$ is not equal to $\Delta\rho_{nlj}/\rho_{nlj}$.

2. For identical nuclei in two ions

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta\alpha}{1+\alpha} = \frac{1}{1+\alpha}\sum_{nlj}\alpha_{nlj}\frac{\Delta\alpha_{nlj}}{\alpha_{nlj}}, \quad \alpha = \sum_{nlj}\alpha_{nlj}. \tag{1}$$

It is shown in a number of papers[5–8] that, with fair accuracy (~1% for $M1$ transitions,[6] ~5% for some others[7,8]) the ICC for electron subshells that differ only in the principal quantum number n, at transition energies considerably exceeding the internal-conversion threshold, are proportional to the electron densities near the nucleus:

$$\alpha_{nlj} \cong k_{lj}\rho_{nlj} \tag{2}$$

(k is the proportionality coefficient). On this basis, frequent use is made of the assumption that the chemical changes $\Delta\alpha_{nlj}$ are also proportional to the changes of the electron densities. In this case

$$\frac{\Delta\lambda}{\lambda} = \frac{1}{1+\alpha}\sum_{nlj}\alpha_{nlj}\frac{\Delta\rho_{nlj}}{\rho_{nlj}}. \tag{3}$$

From (2), however, it follows that

$$\Delta\alpha_{nlj} = k_{nlj}\Delta\rho_{nlj} + \Delta k_{nlj}\rho_{nlj}, \tag{4}$$

where, with several-percent accuracy, all $k_{nlj} = k_{lj}$. Since the main contribution to the total ICC is made by the inner electron shells, it can be seen from (4) that changes even in the fourth or fifth significant figures of the coefficients k_{nlj} for deep inner subshells, changes that have no effect on the accuracy (several percent) of relation (2), can make a large contribution to the change of the total ICC and to the change $\Delta\lambda/\lambda$. From a comparison[1] of the measured $\Delta\lambda/\lambda$ and that calculated from Eq. (3) for ^{119m}Sn in SnO_2 and in β-Sn it follows that such "chemical" changes of the proportionality coefficients k_{nlj} do indeed take place.

3. To obtain more accurate theoretical values of $\Delta\lambda/\lambda$ we need direct calculations of the ICC for different tin ions. They can be performed by using the existing programs.[9] The problem is, however, that we need to have very accurate values of the ICC. The expected changes of $\Delta\lambda/\lambda$ are of the order of 10^{-4}. Therefore the ICC for the K and L shells, whose contribution to the total ICC is in this case 84% (as calculated by the program of Ref. 9) must be calculated accurate to at least five significant figures. In the tables only three figures are given for the ICC.[11,12] A higher accuracy of the absolute values of the ICC has no physical meaning, in view of the approximate character of the physical models used in the calculations. When ICC are calculated for ions with an aim at determining $\Delta\lambda/\lambda$, the difficulty can be circumvented by calculating the ICC with more significant figures within the framework of the existing models, but with physical meaning given only to relative quantities, to the differences

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TABLE I. Calculated ICC α_{nlj} for the 65.3-keV $M4$ transition in the ^{119}Sn nucleus for the neutral tin atom, and the changes $\Delta\alpha_{nlj}$ and the ratio of the changes $(\Delta\alpha_{nlj}/\alpha_{nlj})/(\Delta\rho_{nlj}/\rho_{nlj})$ for the Sn^{4+} ion (relative to the neutral atom).

Electron subshell	Sn^0 $5s^2p^2_{1/2}$	Sn^{4+}	
	α_{nlj}	$\Delta\alpha_{nlj}$	$(\Delta\alpha_{nlj}/\alpha_{nlj})/(\Delta\rho_{nlj}/\rho_{nlj})$
K	1704.952	1.488	63.22
L_1	814.838	0.374	10.98
L_2	181.596	0.096	5.22
L_3	1838.334	0.687	3.68
M_1	189.316	0.081	2.77
M_2	40.992	0.019	2.32
M_3	435.310	0.182	1.77
M_4	6.971	0.003	1.53
M_5	13.274	0.005	1.27
N_1	39.864	0.125	1.09
N_2	7.824	0.046	1.08
N_3	82.664	0.582	1.02
N_4	1.067	0.037	1.00
N_5	2.021	0.075	1.00
O_1	4.472	−4.472	1.00
O_2	0.522	−0.522	1.00
Sum	5364.017	−1.194	–

$\Delta\alpha_{nlj}$ or the relative changes $\Delta\alpha_{nlj}/\alpha_{nlj}$ (under the condition that the ICC are calculated for the compared ions within the framework of the same physical model).

Band and Trzhaskovskaya[12] published the values of α_{nlj} for this transition for the neutral atom and for different tin ions, accurate to four significant figures. This accuracy is insufficient to obtain the theoretical $\Delta\lambda/\lambda$. The ICC for the ions were therefore calculated by the same program[9] with the same number of figures, so that when the Sn^{4+} ion (which has no valence electrons) is compared with the neutral tin atom SnO the changes $\Delta\alpha_{nlj}$ can be observed for each subshell. The result of these calculations of the ICC for SnO and Sn^{4+} are listed in Table I. It can be seen that a very large contribution to the change $\Delta\alpha$ of the total ICC is made by the electrons of the deep inner shells (K, L). It is seen from the last column of the table that this contribution is many times larger than that calculated under the assumption that $\Delta\alpha_{nlj}/\alpha_{nlj} \cong \Delta\rho_{nlj}/\rho_{nlj}$.

The reasons for this are the high multipolarity and the low energy of the transition. The internal conversion takes place in a small volume at the center of the atom.[13] The approximate proportionality of the ICC to the electron density near the nucleus is due to the fact that the main contribution to the ICC is made by the atom region at the nucleus itself. With increasing multipolarity, the volume in which the ICC if formed and the contribution made to the ICC by the more remote regions of the atom increase.[13] Also contributing to this are the increased wavelength of the converted electron when the transition energy is decreased (this manifests itself particularly strongly for conversion on K electrons). As a result, the changes of the ICC are no longer proportional to the changes of the electron densities near the nucleus. The relative changes $\Delta\alpha_{nlj}/\alpha_{nlj}$ for the deep inner shells are close to the relative changes of the internal-conversion electron wavelengths (owing to the change in the binding energy). This shows that the increase of the ICC on the K, L, and M electrons in the ions is due to a unique volume effect—to the increase of the volume in which the ICC is formed.

Figure 1 shows, as functions of the tin-ion charge,

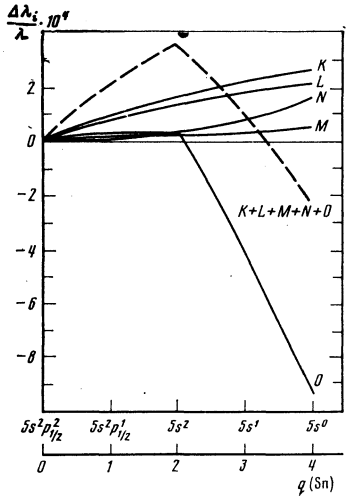

FIG. 1. Contributions to $\Delta\lambda/\lambda$ from the K, L, M, N, and O shells for different tin ions as functions of the ion charge q(Sn) (relative to a neutral atom with valence-electron configuration $5s^2p^2_{1/2}$).

the calculated contributions to $\Delta\lambda/\lambda$ from the K, L, M, N, and O electrons upon successive detachment of the valence electrons from the atom (relative to the neutral tin atom). It is seen that a very large contribution to $\Delta\lambda/\lambda$ is made by the electrons of the deep inner shells. A calculation with allowance for this contribution of the change of $\Delta\lambda/\lambda$ for ^{119m}Sn in SnO_2 compared with β-Sn (assuming that the respective occupation numbers n_{5s} and n_{5p} of the valence $5s$ and $5p$ subshells are 0.60 and 1.30 for SnO_2 and 1.8 and 2.2 for β-Sn, Ref. 4) yielded a value $\Delta\lambda/\lambda = -1.2\times 10^{-4}$. It differs from the measured one only by approximately one mean squared error of the experimental result.

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