

Emergence of a hole through the electron cloud of an atom and energy of the x-ray line excited during a conversion cascade

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An energy shift of the $K\alpha_1$ x-ray line of Te excited in a conversion cascade has been detected experimentally. The effect is attributed to residual vacancies in the electron cloud of the atom when the recombination times are longer than the lifetime of the intermediate state of the nucleus.

1. During electron capture¹ and internal conversion,² the hyperfine structure components of an excited atomic state may be filled in a nonstatistical manner. As a result, the energies of the x-ray lines accompanying these processes will be shifted by an amount proportional to the magnetic moment of the nucleus. This effect has been observed experimentally^{1,2} and has been exploited to measure the magnetic moments of nuclear states.³ In experiments of this type it is very important to correctly allow for several other mechanisms which also shift x-ray lines: chemical shifts, isotopic and isomer shifts,³ "shaking" of electrons, etc.⁴

2. In the present letter we wish to call attention to yet another mechanism which gives rise to an energy shift of x-ray lines excited in cascade processes. This mechanism stems from the finite relaxation time of an atomic shell.

As an example we consider a cascade of two successive K -conversion transitions in an isolated atom. The first of these transitions causes the nucleus to go into an intermediate state with a lifetime τ_i , and a hole is produced in the K shell. This hole moves very rapidly to the outer shell. Initially the hole advances by means of radiative transitions with times 10^{-17} – 10^{-15} s; later, Auger processes (with times 10^{-14} – 10^{-15} s) assume the leading role and create additional vacancies, which also move to the periphery of the atom. If the atom is in a condensed medium, the hole states recombine; their lifetime τ_p depend strongly on the properties of the atomic surroundings. In metals these times are short ($\tau_p \lesssim 10^{-12}$ s), while in insulators they are long⁵ ($\tau_p > 10^{-8}$ s).

If $\tau_i < \tau_p$, the second conversion transition thus occurs while holes are present in the electron cloud of the atom; the effect is to shift the excited K line by an amount which depends on the particular states having the vacancies.

3. For an experimental study of this effect we selected the isomer ^{125m}Te , which decays in a cascade of two conversion transitions to the ground state with an intermediate state of lifetime $\tau_i = 1.48 \times 10^{-9}$ s. The measurements were carried out in a crystal-diffraction spectrometer by the method described in Refs. 3 and 6. Two experiments were carried out. In the first, we measured the shift of the $K\alpha_1$ line of Te, which accompanies the conversion cascade in the isomer ^{125m}Te , taken in the form of a

metal, with respect to the $K\alpha_1$ line excited by photoionization in a metallic Te sample. The experimental shift turned out to be

$$\Delta E_1^{K\alpha_1} ({}^{125m}\text{Te}_{\text{met}} - \text{Te}_{\text{met}}^{\text{photo}}) = -43.3 \pm 2.1 \text{ meV.} \quad (1)$$

In the second experiment, the isomer ${}^{125m}\text{Te}$ and a Te sample excited by photoionization were taken in the form of the dioxide (TeO_2), which is an insulator. The corresponding experimental shift was found to be

$$\Delta E_2^{K\alpha_1} ({}^{125m}\text{TeO}_2 - \text{Te}^{\text{photo}}\text{O}_2) = -64.9 \pm 2.2 \text{ meV.} \quad (2)$$

Several effects contribute to shifts (1) and (2): the hyperfine shift, which is the most important component, and isotopic and isomer shifts. Since these are nuclear effects, they do not depend on the chemical state of the substance and are thus identical in the two experiments. There is no chemical shift in the two cases, since the samples being compared were taken in identical chemical forms. The only difference was that in the second experiment (with the ${}^{125m}\text{Te}$ isomer in the form of TeO_2) the recombination time of the holes which emerge through the electron cloud was longer than the lifetime of the compound state of the nucleus, $\tau_p \gg \tau_i$ (in contrast with the condition $\tau_p \ll \tau_i$, which held in the first experiment). In this second experiment we should therefore see an additional energy shift due to the presence of holes in the outer shells of the atom.

The difference

$$\delta(\Delta E^{K\alpha_1}) = \Delta E_2^{K\alpha_1} - \Delta E_1^{K\alpha_1} = -21.6 \pm 3.0 \text{ meV} \quad (3)$$

can therefore be interpreted as that component of the shift of the $K\alpha_1$ line which is caused by those x-ray transitions that occur when there is a vacancy in the electron cloud of a Te atom whose nucleus decays in an insulator.

4. Since shift (3), due to the emergence of holes through the cloud, is comparable in magnitude to hyperfine shifts (1), a study of this effect is important for eliminating additional shifts both in measurements of nuclear magnetic moments and in other experiments involving fine measurements of the shifts of x-ray lines. On the other hand, this shift may also be of independent interest, since it can in principle be used to study relaxation processes in atoms.

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⁵K. Siegbahn (editor), Alpha-, Beta-, and Gamma-Ray Spectroscopy, North-Holland, Amsterdam, 1965.

⁶O. I. Sumbaev, Usp. Fiz. Nauk **128**, 281 (1978) (*sic*).