MOSSBAUER EFFECT ON Np^{237}

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The isotope Np^{237} has a Mossbauer level with energy 59.6 keV and a lifetime 6.3×10^{-8} sec. The most convenient source of Mossbauer radiation is Am^{241}, the α decay of which leads to the formation of this isomer of Np^{237}. The Mossbauer spectra corresponding to different Am compounds yield information on the stabilization of Np within a time $\sim 10^{-7}$ sec following the α decay of the Am. What is remarkable is that the isomer shifts of the lines reach several cm/sec. The increased sensitivity of the shifts to the electron surrounding can serve as an effective means for the investigation of the state of Np in crystalline substances.

We investigated the Mossbauer effect on Np using different sources and absorbers. The sources were AmO_2, $AmO_2(NpO_2)$ with AmO_2 isomorphically introduced into the NpO_2 lattice, AmF_3, and metallic Am introduced into a thorium lattice - Am(Th). The absorbers were NpO_2 and Np_3O_8. The parameters of the crystal lattices of the Am and Np compounds were determined by x-ray structure analysis and obtained from the corresponding known data.

The Mossbauer spectra were measured at 77°K and at velocities up to $\pm$ 10 cm/sec, using apparatus of the electrodynamic type. The γ radiation was registered with an NaI(Tl) crystal 2 mm thick. The registered part of the γ radiation corresponded to the photopeak from 59.6 -keV γ rays. The spectrum was recorded with a 400-channel analyzers. In order to take into account the background due to the γ radiation of Pa^{233} (the Np^{237} decay product) the number of counts from the absorber and from the source with the absorber were compared during the same active time of analyzer operation.

The absorber used in the investigation of the Mossbauer emission spectra was NpO_2. The AmO_2 source yields several lines, one of which is close to zero velocity [1]. The isomer shift of this line, as measured by us, was 0.7 ± 0.1 mm/sec, in good agreement with the ~ 0.7 mm/sec obtained in [2]. The AmO_2 source was used to study the magnitude of the effect (line with $\delta = 0.77$ mm/sec) as a function of the thickness of the NpO_2 absorber. It was found that a saturation of the effect, $\sim 1\%$, is attained at a thickness ~ 150 mg/cm^2. Therefore the use of thicker absorbers is not advisable.

The Mossbauer spectrum of the $AmO_2(NpO_2)$ source revealed the line at $v \simeq 0$ which was observed in other investigations [3, 4]. But in addition to this line, a line at 4.0 ± 0.4 mm/sec was also observed. Whereas the effect corresponding to the first line was equal to 1%, the effect of the second was $\sim 0.15\%$. No other lines were observed, accurate to 0.03%, in the remaining region of the investigated velocities (up to 10 cm/sec).

According to [1], AmF_3 has at 4.2°K a characteristic line at $v = -4.4$ cm/sec. We observed at 77°K, besides this line, a more intense line near $v = 0$.

The Am(Th) source with an NpO_2 absorber gives the narrowest of the known lines with half-width Γ_{exp} (width at half the height) equal to 1.7 ± 0.2 mm/sec, with a shift

δ = 3.4 ± 0.2 mm/sec. It is estimaged that the half-width of the Am(Th) source line is approximately 0.4 mm, i.e., it exceeds the natural width Γ_{nat} = 0.036 mm/sec by approximately 10 times [5].

Measurements with the Am(Th) source and an Np_3O_8 absorber have shown that the spectrum contains a line with an isomer shift δ = -2.1 cm/sec, the parameter of the quadrupole splitting amounting to 1/4eqQ = 1.4 ± 0.2 cm/sec. It is interesting to note that the same line has a larger quadrupole splitting at 4.2°K, namel 1/4eqQ = 2.58 ± 0.03 cm/sec. Another line, with a shift δ = -1.55 cm/sec was also found in [1], having both quadrupole and magnetic splitting. This line was not observed by us at 77°K, accurate to 0.03%.

On the basis of the obtained data we can establish a correlation between the isomer shifts and the charge states of the neptunium produced after the α decay of americium. The state Np^0 corresponds to a shift with relatively low velocity, as follows from the position of the Am(Th) source line. The shift for Np^{+4} apparently also falls in the region near v = 0, and is observed in the sources AmO_2 and $AmO_2(NpO_2)$.

Two states of neptunium arise in the AmF_3 source: Np^{+4} with δ = 0 and Np^{+3} with δ = -4.1 cm/sec. On going from Np^{+4} to a state with a larger charge (Np_3O_8), the shift becomes positive and equal to 2.44 cm/sec. Indeed, the position of the -2.1 cm/sec line of the Np_3O_8 absorber relative to the line of the $AmO_2(NpO_2)$ source, at v = 0, is at 2.1 + 0.34 = 2.44 cm/sec; the sign of the shift changes here and account is taken of the line shift of the Am(Th) source used in the experiments with the Np_3O_8.

The results do not contradict the data of [1], where the regions of the isomer shifts were determined for various charge states of neptunium at 4.2°K. According to these data, the transition from Np^{+3} to Np^{+6} is connected with the successive change of the shifts; in addition, the change of the shift on going from Np^{+3} to Np^{+4} is maximal. If allowance is made for the screening action of the 5f electrons on the 6s electrons, then it follows from the first fact that the Np^{237} nucleus is less deformed in the excited state than in the ground state ($\Delta\alpha/\alpha$ is negative).

In the state Np^{+3}, compared with Np^{+4}, there remains an extra 6d or 5f electron. The screening action of the 6d electron on the 6s electrons is larger than the action of the 5f electron. Therefore the maximum shift on going from Np^{+3} to Np^{+4} apparently shows that the charge state of Np^{+3} corresponds to an electron configuration $6f^36d$.

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