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DEFECT $^{119}$Sn ATOMS AFTER NUCLEAR DECAYS AND REACTION IN SnSb AND SnTe

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Résumé. — On a étudié par spectroscopie d’émission le site des atomes $^{119}$Sn créés par capture électronique et réaction protonique dans SnSb et SnTe. Dans Sn$^{119}$Sb et Sn$^{120}$mTe, les atomes $^{119}$Sn ont été trouvés dans les sites de Sb ou Te des matrices. Les atomes de recul $^{119}$Sb créés par réaction protonique, ont été trouvés uniquement dans les sites de Sb dans SnSb alors que dans SnTe ils sont distribués dans les sites de Sn et Te.

Abstract. — The lattice position of $^{119}$Sn atoms after EC decays and a proton-reaction was studied in SnSb and SnTe by emission spectroscopy. In Sn$^{119}$Sb and Sn$^{120}$mTe, defect $^{119}$Sn atoms were found in the Sb or Te site of the matrices. The $^{119}$Sb recoil atoms after the proton reaction were found only in the Sn site of SnSb, but they were distributed between both the Sn and Te sites of SnTe.

The defect structures of $^{119}$Sn atoms produced by nuclear decays and reaction have been investigated in the matrices of SnSb and SnTe as a part of Mössbauer emission studies on $^{119}$Sn with $^{119}$Sb as the source nuclide [1-4]. The nuclear processes studied were the following ones characterized by different recoil energies ($E_R$) relative to the displacement energy in solid ($E_D$) :

(a) $^{119}$Sb $\rightarrow$ $^{119}$Sn ($E_R \ll E_D$)
(b) $^{119}$mTe $\rightarrow$ $^{119}$Sb $\rightarrow$ $^{119}$Sn ($E_R \approx E_D$)
(c) $^{120}$Sn(p, 2n) $^{119}$Sb $\rightarrow$ $^{119}$Sn ($E_R \gg E_D$).

The Mössbauer emission spectra of $^{119}$Sn arising from $^{119}$Sn were measured in (a) and (b) on the SnSb and SnTe sources labeled with $^{119}$Sb or $^{120}$mTe, and in (c) on the samples irradiated by protons. A preliminary report on the proton-irradiated SnTe has been already made [5].

Tin metal depleted in $^{119}$Sn($^{120}$Sn 98.39 %, $^{119}$Sn 0.39 % referred to hereafter as $^{120}$Sn) was employed in preparing the sources to minimize the resonant self-absorption of the Mössbauer γ-rays. Irradiation of the $^{120}$SnSb and $^{120}$SnTe samples were carried out with 15 MeV protons on a water-cooled aluminum plate under a helium atmosphere. The irradiated powder samples gave no visible indication of melting. The emission spectra were measured at 78 K against a BaSnO$_3$ absorber (0.9 mg $^{119}$Sn/cm$^2$) at the same temperature. Measurements on the $^{120}$Sn$^{119}$mTe samples were started after a radioactive equilibrium has been established between $^{120}$mTe and $^{119}$Sn.

The $^{120}$Sn$^{119}$Sn sources gave an emission line with an isomer shift of 2.43 ± 0.03 mm/s relative to BaSnO$_3$ at 78 K, as may be seen in figure 1a. Comparison of the isomer shift with that of the absorption line of the same compound (2.79 ± 0.03 mm/s) indicates that the $^{119}$Sn atoms arising from $^{119}$Sb in the SnSb matrix are in an electronic state different from that of the tin atoms in the normal Sn site of SnSb. Since the recoil energy associated with the EC decay of $^{119}$Sn to $^{119}$Sn is much smaller (1.4 eV) than the displacement energy, no displacement of the $^{119}$Sn atoms from the original site of $^{119}$Sn is expected. Therefore, the observed emission line is attributed to the defect $^{119}$Sn atoms in the Sn site of the SnSb matrix.

As the recoil energy of the $^{119}$mTe $\rightarrow$ $^{119}$Sn is estimated to have a distribution with a minimum value below the displacement energy and a maximum exceeding it, partial displacement of the decaying atoms is expected in the successive decays, $^{119}$mTe $\rightarrow$ $^{119}$Sb $\rightarrow$ $^{119}$Sn. In fact, two lines with isomer shifts of 2.24 ± 0.05 mm/s and 3.3 ± 0.2 mm/s were observed in the emission spectra of $^{120}$Sn$^{119}$mTe (Fig. 1b). The latter line is attributed to $^{119}$Sn in the Sn site of SnTe, since the isomer shift is close to that of the absorption line of the same compound (3.54 ± 0.05 mm/s). The former line is consequently attributed to $^{119}$Sn resting in the Te site.

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On the basis of these results together with the data reported previously [2, 3], a systematics was established between the isomer shift of defect and normal $^{119}$Sn atoms and the electronegativity of the nearest neighbor atoms in metals and binary compounds of Sn, Sb and Te. In these systems the isomer shift is determined to the first approximation by the nearest-neighbor atoms and increases with an increase in the electronegativity of the nearest-neighbor atoms.

From the isotopic composition of tin and the energy of protons the main reaction leading to $^{119}$Sb in the proton-irradiation is estimated to be the $^{120}$Sn(p, 2 n)$^{119}$Sb. The accompanying recoil energy is so large ($\sim 10^2$ keV) that all the $^{119}$Sb atoms are knocked out from the original site of $^{120}$Sn. Since the following EC decay of $^{119}$Sb to $^{119}$Sn brings about no atomic displacement, the lattice position of $^{119}$Sb is considered to have been the same as that of its daughter $^{119}$Sn, which is estimated from the emission spectra.

As may be seen in figure 2a, the $^{119}$Sn atoms arising from $^{119}$Sb recoiled by the proton-reaction in $^{120}$SnSb gave an emission line with an isomer shift of 2.42 ± 0.03 mm/s. The isomer shift is essentially the same as that of the labeled source $^{120}$Sn$^{119m}$Te. This shows that the $^{119}$Sn atoms and, accordingly, also the $^{119}$Sb atoms were in the Sb site of the matrix.

The emission spectra of proton-irradiated $^{120}$SnTe were composed of two lines with isomer shifts of 2.3 ± 0.1 mm/s and 3.48 ± 0.05 mm/s (Fig. 2b), indicating that the recoil $^{119}$Sb atoms came to rest in two different states. The latter line is attributed to $^{119}$Sn in the normal lattice position of Sn in SnTe, because its isomer shift is the same as that of the absorption line of SnTe within the experimental errors. The former line is attributed to $^{119}$Sn in the Te site (possibly accompanied with some near-by defects), referring to the dominant emission line of the labeled source $^{120}$Sn$^{119m}$Te.

The absorption spectra of the irradiated $^{120}$SnSb and $^{120}$SnTe revealed no radiation damage of the matrix by protons, giving only one line corresponding to $^{119}$Sn in the normal lattice site. This confirms that the emission spectra described above do not reflect the macroscopic radiation effects of protons, but reveal the consequences of the proton reaction leading to $^{119}$Sb.

In the proton-irradiated $^{120}$SnSb, one cannot disregard the possibility that the observed distribution of $^{119}$Sn is the result of local melting of the matrix along the recoil track. In $^{120}$SnTe, however, the idea is not compatible with the following observations. Firstly, the distribution shows a marked change by thermal annealing after irradiation. Secondly, the $^{120}$SnTe samples melted once after irradiation give a different spectrum.

It may be noteworthy that the $^{119}$Sb atoms were found in the two distinct lattice positions of $^{120}$SnTe after recoil by the proton reaction. This shows that...
the SnTe lattice is preserved to a large extent in the vicinity of the $^{119}$Sb atoms and suggests that the $^{119}$Sb atoms have been stabilized at one of the lattice points with a rather unperturbed environment as the result of a replacement collision and the following annealing process.

References