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INVESTIGATION OF CHANGES IN HYPERFINE INTERACTION AND DEBYE-WALLER-FACTOR BY α-RADIATION SELF DAMAGE IN ^{241}Am METAL*

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Résumé.- Les effets de l'auto-irradiation α de ^{241}Am métal ont été étudiés par spectroscopie Mössbauer d'émission. Nous avons maintenu les échantillons à 4,2 K pendant 230 h d'affilée et relevé des spectres Mössbauer toutes les 10 à 20 heures. Il n'y a aucune variation du facteur f, par contre la largeur de raie augmente progressivement pour atteindre sa valeur à saturation après 180 h. Un bref recuit à température ordinaire régénère la largeur initiale. L'augmentation de largeur de raie reflète un changement de l'interaction quadrupolaire lié à la création de défauts. La valeur constante du facteur f indique que la structure cristalline n'est pas radicalement modifiée.

Abstract.- The Mössbauer emission spectrum of ^{241}Am metal was investigated for influences of radiation self damage. Samples were kept continuously for 230 h at 4.2 K and spectra were taken each 10 to 20 h. No change in f-factor was observed, while the linewidth increased monotonically and reached saturation after ∿ 180 h. The original width could be reproduced by a brief warming to room temperature. The increase in width reflects the change in quadrupole interaction due to the creation of lattice defects. The constancy of the f-factor indicates that the basic crystalline structure is retained.

The commonly used source for Mössbauer measurements with ^{237}Np is ^{241}Am which decays into the 60 keV excited state ($T_{1/2}$= 63 ns) by the emission of a 5.5 MeV α-particle. It is astonishing, that the observation of a clear Mössbauer signal is possible some ten nanoseconds after such a violent act as an α-decay which imparts about 100 keV recoil energy to the neptunium nucleus. Aftereffects of the preceeding α-decay are known to exist. Best established is the observation of multiple emission lines in sources like AmO_2 /1/. In the present experiment we look for changes in time of the resonance line of Am metal sources kept at liquid helium temperature for prolonged times. Such changes could be caused by radiation self damage which is known to play a role in measurements of the heat capacity /2/ and the electrical resistivity /3/. Measurements were performed using a 100 mg/cm^2 absorber of NpO_2 at 77 K. A fast pulse detection system /4/ allowed the compilation of precise spectra in short time. Data were least squares fitted to a single Lorentzian modified by a dispersion amplitude of ξ = -0.034 /5/. The recoilfree fraction is proportional to the total area under the resonance curve which was determined by a point by point summation over the whole velocity range after the baseline had been fixed in the least squares procedure. Two different sources of ^{241}Am metal were studied. They were both produced by vacuum evaporation of Am metal onto a Ta disc. They differ in the total amount of Am metal (15.5 mg and 89 mg) and their radioactivity (53 mCi and 306 mCi). Their mass (activity) per unit area, however, is roughly the same (~ 100 mg/cm^2). Figure 1 displays for the 53 mCi source the linewidth, peak height and total area as a function of time. The area remained constant, but the linewidth showed a monotonic increase reaching saturation after ~180 h. After 10 days at 4.2 K the sample was briefly warmed to room temperature, then recooled. Within the limits of error the initial values of width, height and area were reproduced. Similar results were obtained for the 306 mCi sample.
In another, yet unpublished, measurement we found that the resonant lineshape for Am at 4.2 K can be fitted to an unresolved, purely axial quadrupole interaction of $e^2qQ \cong 6$ mm/s ≙ 300 MHz. Such an interaction is expected from the double hcp structure of bulk Am metal /6/. Other sources of line broadening, like a distribution of isomer shifts, will also be present but their size is difficult to estimate. The observed change in linewidth is interpreted as a gradual increase of quadrupole inter-

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action connected with the radiation damage of the Am lattice.

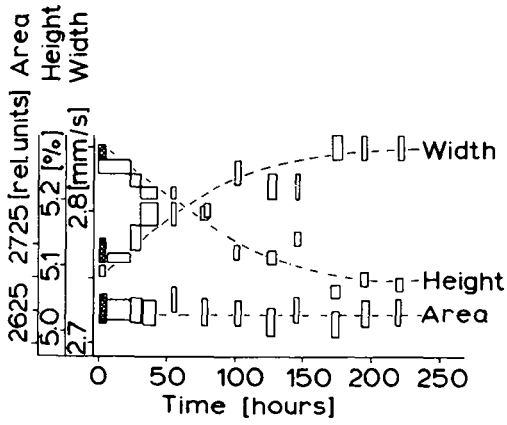

Fig. 1 : Change of spectral parameters with time for the 53 mCi sample of ^{241}Am metal kept at 4.2 K. The cross hatched data were taken at the end of the run after a warm-up to 300 K.

Both, the α-particles of 5.5 MeV and the Np recoil nuclei will primarily create vacancies and interstitials in their passage through the lattice /7/. If located in the vicinity of the emitting Am atom, these lattice defects will give rise to an additional electric field gradient /8/. The primary recoils will travel only ~8 nm and stop in a time short compared to the half life of the 60 keV excited state. They are surrounded mainly by a cloud of vacancies while the interstitials have moved further away by correlated collision processes. The damages remaining in the vicinity of the emitting atom itself could at best contribute to the initial linewidth but should not show a build-up in time. This is different with the general radiation damage inflicted on the Am lattice by both the nuclear recoils and the α-particles. The recoils create damage in high concentration while the α-particles produce vacancies and interstitials over a much larger volume. These damages will be frozen in at helium temperature and cannot easily annihilate together. Their concentration will rise and soon the whole Am lattice is heavily afflicted. Finally the vacancies and interstitials will come close enough together to annihilate even at low temperatures and a stationary concentration of defects will results. The thickness of both Am samples is large compared to the mean stopping distance of either the nuclear recoils or the α-particles (~ 8 μm). Thus the number of damaged sites produced per unit volume will be the same for both samples and the similarity of their behaviour is understandable.

This view of the origin of the time dependent line broadening is further supported by measurements of transport properties /2,3/. In particular, an exponential growth of resistivity with a time constant of 50 h was found. Warming to room temperature nearly but not exactly reproduced the original resistivity. The small amount of remanent radiation damage is at the limit of sensitivity of a Mössbauer measurement.

The constancy of the f-factor with time indicates on a local picture that no diffusion and subsequent trapping at the Np site occur for the interstitials within the lifetime of the 60 keV state. It is known that impurity-interstitial complexes have low vibrational frequency modes causing a reduction in f-factor /9/. One expects little migration at 4.2 K. Furthermore, the electronic structures of neptunium and americium are not very different and the former will not be an efficient trap. In terms of non-local properties one may expect that the crystal structure will severely be destroyed under the influence of self-irradiation. Our result makes an "amorphization" of Am metal rather unlikely, since noticeable differences in the f-factor usually exist between crystalline and amorphous phases /10/. The material may well become microcrystalline under the influence of self-irradiation but retains its basic crystalline structure.

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