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GAMMA-X RAY COINCIDENCE MÖSSBAUER SPECTROSCOPY WITH COBALT CHLORIDE

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Résumé.- A partir d'une source de CoCl₂.2H₂O on a observé un spectre Mössbauer modifié par l'émission intense d'électrons de basse énergie et de rayons X, sans que la cascade Auger ne se produise. Par une méthode de coincidence, on met en évidence une différence entre les paramètres Mössbauer mesurés dans ces conditions et ceux résultant d'expériences classiques.

Abstract.- Using the γ -KX ray coincidence technique, a spectrum influenced by low-energy electrons and X rays of which numbers are different from those in the case of simple measurements has been observed with a source of CoCl₂.2H₂O. The obtained values of parameters are different from those obtained by simple measurements.

Many workers /1-5/ have observed by Mössbauer emission spectroscopy that anormalous charge states are stabilized in insulating materials labelled or doped with ⁵⁷Co. This fact has been interpreted as due to the self-radiolysis of the ligand environment by the low-energy electrons and X rays emitted during the Auger cascade following the electron capture (EC) decay of ⁵⁷Co /2,5/. When a KX-ray is emitted after the EC decay without the K-Auger process, the number of low-energy electrons and X rays originated by the Auger cascade is about half compared with those when it is not emitted /6/. In such a case, by introducing the $\gamma\text{-}KX$ ray coincidence technique, we can observe a spectrum influenced by low-energy electrons and X rays of which numbers are different from those in the case of simple measurements. In the present paper, we report on Y-X ray coincidence Mössbauer measurements on ⁵⁷CoCl₂.2H₂O. The measuring system is very similar to that for the delayedcoincidence Mössbauer experiments /7/.

The observed spectra can be decomposed into four doublets of Fe^{2+} and Fe^{3+} ions /5/. The parameters estimated by the method of least-squares are listed in table I (see Appendix). From the magnitude of isomer shifts and quadrupole splittings, we ascribe the four doublets to ground and excited states of Fe^{2+} and Fe^{3+} , which are represented by Fe^{2+} , Fe^{2+*} , Fe^{3+} and Fe^{3+*} , respectively.

The yield of Fe²⁺ obtained from the coincidence spectrum is almost the same as that obtained from the spectrum without coincidence. This fact means that the ratio of anomalous states formed after the EC decay of ⁵⁷Co to the normal state does not change, though the ratios between the anomalous state change, even when the emission rate of low-energy electrons and X rays goes down to about half. The small values of Λ of Fe $^{2+}$ and Fe $^{3+}$ are caused by good homogeneity of nuclear environment, and does not depend strongly on the emission rate. The values of the yield and Λ of Fe²⁺**x** and Fe³⁺**x** obtained from the spectrum without coincidence are different from obtained from the spectrum with coincidence. This can be probably explained by the difference of the emission rate when the spectra are observed.

We are now working to obtain spectra of sufficiently good statistics with sources of $CoCl_2.2H_2O$, $CoCl_2.6H_2O$ and $CoSO_4.7H_2O$.

	Isomer Shift (mm/s)	Quadrupole Splitting (mm/s)	Without A (mm/s)	Coincidence Yield (%)	With Coincidence	
					(mm/s)	(%)
Fe ²⁺	-1.24	2.39	0.12	36.1	0.01	35.4
Fe ²⁺	-1.20	1.74	0.47	25.2	0.77	35.5
Fe ³⁺ ₹	-0.34	1.20	0.77	34.0	0.53	20.4
Fe ³⁺	-0.25	0.62	0.06	4.7	0.08	8.7

Table I : Mössbauer Parameters

<u>Appendix</u>.- The linewidth of each peak is broader than that expected by taking account of the absorber thickness. Since in the present measurement the line broadening due to the sample thickness and collimation geometry can be neglected, this discrepancy between measured and expected spectra may be supposed to be ascribed to the fact that isomer shifts and quadrupole splittings are distributed around their mean values owing to the time-independent nuclear environmental inhomogeneity and the relaxation phenomena. Therefore, in the present work, we assume that each unfolded component peak of the spectra can be expressed by a convolution of a Gaussian and a funct<u>ion</u> as

$$f(V) = \int_{\infty}^{\infty} \exp\left(-\frac{4\ln^2}{\Lambda^2} v^2\right) h(V-\delta-v) dv,$$

where V is a Doppler shift, Λ a half-width of a Gaussian term which depends on the nuclear environmental inhomogeneity and the relaxation phenomena following the EC decay of ⁵⁷Co, δ an isomer shift, and the function $h(V-\delta-v)$ is an absorption peak expected by taking account of the absorber thickness. A more detailed discussion of the analysis will be presented by one of the authors (T.K.) in a forthcoming paper.

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