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QUADRUPOLE INTERACTION AND ANISOTROPY OF THE MOSSBAUER EFFECT AS DEDUCED FROM OBSERVATIONS ON RESONANCE SCATTERING OF GAMMA QUANTA BY POLYCRYSTALS

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The quadrupole interaction of Sn^{119} nuclei in the lattice of white tin is determined by studying the attenuation of the angular distribution of Mossbauer scattering. The values obtained are $E_Q/\Gamma = 0.58 \pm 0.20$, $\Delta = 0.18 \pm 0.6$ mm/sec at 300°K and $E_Q/\Gamma = 0.82 \pm 0.15$, $\Delta = 0.25 \pm 0.05$ mm/sec at 80°K (Γ is the width of the nuclear level and Δ is the hyperfine splitting). The influence of anisotropy of the Mossbauer effect in crystals on the angular distributions of resonance scattering corresponding to excitation of individual components of the allowed quadrupole doublet is considered theoretically. It is shown that even when the measurements are carried out on polycrystalline samples it is possible in principle to derive from the angular-distribution functions both the magnitude and sign of anisotropy of the effect and the sign of the quadrupole interaction. Experimentally, the influence of the Mossbauer effect has been detected in measurements of the angular distributions for the quadrupole doublet components in the compound $(C_4H_9)_2SnO$.

1. INTRODUCTION

THE discovery of the Mossbauer effect has made it possible, in particular, to determine directly the interaction between a nucleus and the fields surrounding it from the hyperfine structure of the resonance-absorption lines. However, when the interaction is relatively small and the γ-line splitting Δ is smaller than or of the order of the width Γ of the nuclear level, only a broadening of the line, which may be large or small, is observed, and in principle the hyperfine interaction may not be its sole cause. In such cases, more definite information on the magnitude of the interaction can be obtained by measuring the angular distribution of the resonant scattering of γ quanta. The results of such measurements with 23.8-keV γ rays from Sn^{119} on the substances Mg_2Sn and SnO_2 were published by us earlier.[1]

On the other hand, in the case of strong interactions, when the hyperfine structure of the γ line is clearly pronounced ($\Delta \gg \Gamma$) it becomes possible to observe resonant scattering by excitation of individual sublevels in the scattering nuclei situated in the exciting field. As follows from the theory of angular correlations, the angular distribution of resonant scattering for any component of a multiplet, is described when $\Delta \gg \Gamma$ for any polycrystalline isotropic sample by the expression for the "hard core".[2,3] A specific feature of experiments on Mossbauer scattering, compared with ordinary γ-γ correlations, is that the angular distributions of the resonant scattering may be influenced by effects connected with the dynamics of the crystal lattice, since the Debye-Waller factor enters in a significant manner in the description of the process. It is known[4-7] that its magnitude can be different in different directions relative to the crystallographic axes, and that this anisotropy can reach appreciable values.[7] Thus, even in the case of random orientation of the microcrystals in the polycrystalline sample, the latter may have anisotropic properties capable of influencing the angular distribution of the resonant Mossbauer scattering.

Karyagin[8] has shown that the anisotropy of the Debye-Waller factor and accordingly of the probability f′ of recoilless γ-quantum absorption should cause a noticeable redistribution of the intensities of the components of the quadrupole multiplet (in particular, the doublet in the case of Sn^{119}) in the resonant-absorption spectra. It is natural to propose that the form of the angular distributions of resonant scattering will be no less strongly influenced by the anisotropy f′, for individual components of the multiplet, than the intensities of the components.

In addition to continuing the previously started[1] measurements of small quadrupole interactions of Sn^{119} nuclei in crystals, we have carried out in this work theoretical and experimental investigations of the influence of the anisotropy of f′ on the angular distributions of resonant scattering by individual components of the quadrupole duplet in crystals of tin compounds. From the attenuation of the anisotropy of the angular distribution of the resonant scattering we have determined the quadrupole interaction of the Sn^{119} nuclei in the crystal lattice of white tin (β-Sn). Measurements on the individual components were made with SnO and $(C_4H_9)_2SnO$, the absorption spectra of which are doublets with large values of Δ. In the latter case a noticeable distortion of the angular-distribution function was noted, apparently connected with the large anisotropy of f′ in this substance.

2. THEORY

In this section we present a short summary of the necessary information from the theory of perturbed angular correlations, and also the results of calculations of the influence of the anisotropy of the recoilless γ-quantum absorption probability on the angular-distribution function of resonant Mossbauer scattering.

The angular distribution of nuclear resonant scattering of γ quanta is described by the formulas of the general theory of angular correlations.[2, 3, 9] In the presence of quadrupole interaction between the nucleus and the intracrystalline fields, it is written for a polycrystalline isotropic scatterer (without account of the anisotropy of f′) in the form

$$W(\theta)=\sum_k A_k G_k P_k(\cos\theta), \tag{1}$$

where G_k are the angular-distribution attenuation coefficients, determined by the interaction of the nucleus in the intermediate (excited) state, and P_k are Legendre polynomials.

We confine ourselves to the case of the transition

$$J_a={}^1/_2\xrightarrow{L_1=1}J_b={}^3/_2\xrightarrow{L_2=1}J_c={}^1/_2, \tag{2}$$

which is realized in Sn^{119}, and also in Fe^{57}. For such a transition, the angular distribution function is[3]

$$W(\theta)=1+0.25G_2P_2(\cos\theta), \tag{3}$$

and in the case of interaction between the nucleus and a static axially-symmetrical electric-field gradient we have[2]

$$G_2=\frac{1}{5}\left[1+\frac{4}{1+E_Q^2/\Gamma^2}\right], \tag{4}$$

where $E_Q = {}^1/_2 QV_{zz}$ is the level splitting and Γ its real width; Q is the quadrupole moment and V_{zz} the tensor component of the electric field gradient. Generally speaking, the G_k depend on the symmetry of the field, but for $J_b = {}^3/_2$ there is no such dependence.[10] It is seen from (4) that G_2 is the most sensitive to the magnitude of the interaction when $E_Q/\Gamma \sim 1$, i.e., in the region where the hyperfine structure of the resonant-absorption spectra is poorly resolved.

In the case of strong quadrupole interaction ($E_Q/\Gamma \gg 1$) the value of G_2 tends to a nonvanishing limiting value $G_{2\min} = 0.2$ (for $J_b = {}^3/_2$), and (3) goes over into the expression

$$W(\theta)=1+0.05P_2(\cos\theta), \tag{5}$$

called the "hard core."

In the case of large splitting in the scatterer, using a source with a single line, we can observe resonant scattering by excitation of individual sublevels of the intermediate state. In general form the angular distribution for this case (quadrupole interaction in the intermediate state) is written (using Alder's notation[3]) in the following manner:

$$W(\mathbf{k}_1,\mathbf{k}_2)=\sum_{\substack{M_a,M_c\\ m_1,m_2}}\left|\sum_{M_b}C^{J_aM_a}_{J_bM_bL_1M_1}C^{J_bM_b}_{J_cM_cL_2M_2}\times D^{L_1}_{M_1m_1}(\mathbf{k}_1)D^{L_2}_{M_2m_2}(\mathbf{k}_2)\right|^2, \tag{6}$$

where in the intermediate state the summation over the indices M_b is carried out only for two values, equal to $+J_{bz}$ and $-J_{bz}$ ($\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the incident and scattered photons).

For a single-crystal scatterer, after performing the foregoing operations, the angular distributions in the coordinate system fixed in the crystal take the form

$$W(\mathbf{k}_1,\mathbf{k}_2)=1+\frac{1}{2\sqrt{5}}Y_{20}(\mathbf{k}_1)+\frac{1}{2\sqrt{5}}Y_{20}(\mathbf{k}_2)+\frac{1}{20}Y_{20}(\mathbf{k}_1)Y_{20}(\mathbf{k}_2) \tag{7a}$$

for the transition

$$|M_a|={}^1/_2\xrightarrow{L_1=1}|M_b|={}^3/_2\xrightarrow{L_2=1}|M_c|={}^1/_2$$

and

$$W(\mathbf{k}_1,\mathbf{k}_2)=1-\frac{1}{2\sqrt{5}}Y_{20}(\mathbf{k}_1)-\frac{1}{2\sqrt{5}}Y_{20}(\mathbf{k}_2)+\frac{1}{20}Y_{20}(\mathbf{k}_1)Y_{20}(\mathbf{k}_2) \tag{7b}$$

for the transition

$$|M_a| = {}^1/_2 \xrightarrow{L_1=1} |M_b| = {}^1/_2 \xrightarrow{L_2=1} |M_c| = {}^1/_2.$$

For a polycrystalline isotropic sample the angular distributions are obtained by averaging (7a) and (7b) respectively over the orientation of the systems of the system of principal axes of the electric-field gradient tensor. Here, as expected from general symmetry considerations, the angular distributions for both components are the same and coincide with the "hard core" (5), where θ is the angle between the vectors k_1 and k_2.

As already noted in the introduction, the use of the existing theory of angular correlations for the description of the angular distribution of the Mossbauer scattering is valid in principle only under the assumption that the probability of the Mossbauer effect does not depend on the direction of propagation of the γ quanta relative to the crystallographic axes, an assumption not satisfied in the general case.

The corresponding quantitative calculation was performed by us for a polycrystalline scatterer with axially-symmetrical microcrystals. The transition (2) was considered in the following cases: 1) $E_Q/\Gamma \lesssim 1$, which corresponds to simultaneous excitation of both components of the doublet, and 2) $E_Q/\Gamma \gg 1$—for separate excitation of the components, and for two methods of registration of the scattered radiation: with the aid of an ordinary nonresonant detector and with the aid of a detector registering only recoilless-scattered γ quanta (resonant detector). It was assumed, just as in [8], that the angular dependence of the recoilless-absorption probability can be represented in the form

$$f'(\vartheta) = a_0 + a_2\bar{P}_2(\cos\vartheta) + \ldots, \tag{8}$$

where $\bar{P}_2(\cos\vartheta)$ is a normalized Legendre polynomial and ϑ is the angle between the direction of the quantum and the principal axis c of the crystal.

A. Weak splitting, nonresonant detector. In this case the anisotropy of f′ is taken into account only for the absorption process. The angular-correlation function in the presence of quadrupole interaction in the single crystal is of the form (Formula (11) of [3])

$$W(\mathbf{k}_1, \mathbf{k}_2) = \sum_{k_1k_2\mu} A_{k_1k_2} G_\mu{}^{k_1k_2} \frac{1}{(2k_1+1)^{1/2}(2k_2+1)^{1/2}} \times Y_{k_1\mu}(\mathbf{k}_1) Y^*_{k_2\mu}(\mathbf{k}_2). \tag{9}$$

Multiplying (9) by (8) and averaging over all orientations of the microcrystals, we obtain after substituting the concrete values of the moments

$$W(\theta) = 1 + \frac{\sqrt{5}}{56}\frac{a_2}{a_0}P_2(\cos\theta) + \frac{1}{4}\left(1 - \frac{\sqrt{5}}{14}\frac{a_2}{a_0}\right)G_2P_2(\cos\theta). \tag{10}$$

We see from this expression that the angular distribution for the total transition is not very sensitive to the degree of anisotropy.

B. Strong splitting, nonresonant detector. Using formulas (7a) and (7b), we obtain in similar fashion the angular distributions for the individual components:

$$W(\theta) = 1 + \frac{7 + 16\sqrt{5}\,a_2/a_0}{14(10 + \sqrt{5}\,a_2/a_0)}P_2(\cos\theta) \tag{11a}$$

or the transition $|{}^1/_2| \xrightarrow{1} |{}^1/_2| \xrightarrow{1} |{}^1/_2|$ and

$$W(\theta) = 1 + \frac{7 - 12\sqrt{5}\,a_2/a_0}{14(10 - \sqrt{5}\,a_2/a_0)}P_2(\cos\theta) \tag{11b}$$

for the transition $|{}^1/_2| \xrightarrow{1} |{}^1/_2| \xrightarrow{1} |{}^1/_2|$.

Figure 1 and Table I show the values of the coefficients of the Legendre polynomials in expressions (11) for an angular distribution of the form

$$W(\theta) = 1 + A_2P_2(\cos\theta) \tag{12}$$

as functions of the degree of anisotropy the quantity f′, equal to

$$\varepsilon = [f'(\pi) - f'(\pi/2)]/[f'(\pi) + f'(\pi/2)]. \tag{13}$$

Table I lists also the ratios of the intensities I of the doublet components in the resonance-absorption spectrum, calculated from the formula given in Karyagin's paper.[8] It turns out that the angular distributions for the individual components are much more sensitive to the value of the anisotropy

Table I. Values of the coefficients of the Legendre polynomials in the angular-distribution functions (12) for individual components of the doublet and component intensity ratios in the absorption spectra as functions of the anisotropy of the Mossbauer effect probability in the crystal

ε	A_2		$1-\frac{I(^3/_2)}{I(^1/_2)}$	ε	A		$1-\frac{I(^3/_2)}{I(^1/_2)}$
	$\|{}^1/_2\| \rightleftarrows \|{}^3/_2\|$	$\|{}^1/_2\| \rightleftarrows \rightleftarrows \|{}^1/_2\|$			$\|{}^1/_2\| \rightleftarrows \|{}^3/_2\|$	$\|{}^1/_2\| \rightleftarrows \rightleftarrows \|{}^1/_2\|$	
—1.0	—0.071	0.123	0.18	0.2	0.065	0.026	—0.06
—0.8	—0.050	0.113	0.15	0.4	0.113	—0.003	—0.13
—0.6	—0.028	0.100	0.12	0.6	0.149	—0.040	—0.22
—0.4	0.004	0.086	0.09	0.8	0.189	—0.087	—0.34
—0.2	0.022	0.070	0.05	1.0	0.232	—0.152	—0.50
0.0	0.050	0.050	0.00				

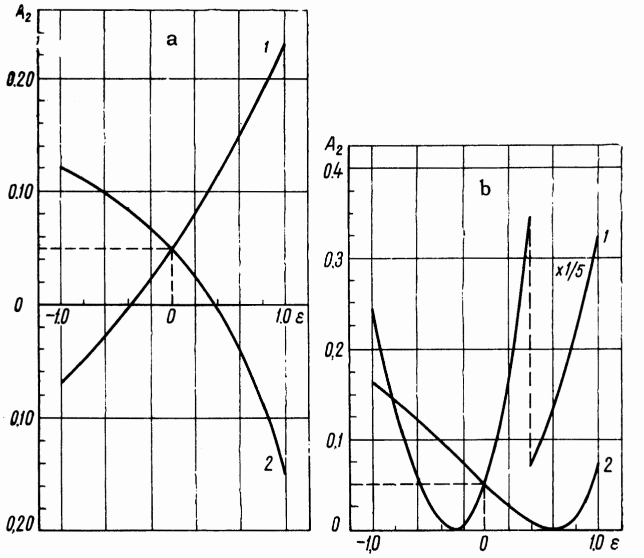

FIG. 1. Dependence of the coefficients of angular distribution of Mossbauer scattering by individual components of the quadrupole doublet on the anisotropy of f′: a—using a nonresonant detector, b—a resonant detector (polycrystalline scatterer). Curves: 1—transition $|{}^1/_2| \rightleftarrows |{}^3/_2|$, curves 2—transition $|{}^1/_2| \rightleftarrows |{}^1/_2|$.

than the angular distribution for the complete transition. Moreover, they are essentially different, and in the case of sufficiently large values of ε (> 0.3) this difference is so large that it becomes possible to determine, not only the absolute value of ε but also its sign and the sign of the quadrupole interaction, something impossible in principle with measurements of the component intensity ratio only (without additional measurements with partially oriented sample[8]).

C. Strong splitting, resonant detector. In this case the anisotropy f′ must be taken into account both for absorption and for emission of the γ quantum. As a result we arrive at the expressions

$$W(\theta) = 1 + \frac{5(1+3\sqrt{5}\,a_2/a_0)^2}{(10+\sqrt{5}\,a_2/a_0)^2} P_2(\cos\theta) + \frac{180(a_2/a_0)^2}{49(10+\sqrt{5}\,a_2/a_0)^2} P_4(\cos\theta) \qquad (14a)$$

for the transition $|{}^1/_2| \xrightarrow{1} |{}^3/_2| \xrightarrow{1} |{}^1/_2|$ and

$$W(\theta) = 1 + \frac{5(1-\sqrt{5}\,a_2/a_0)^2}{(10-\sqrt{5}\,a_2/a_0)^2} P_2(\cos\theta) + \frac{180(a_2/a_0)^2}{49(10-\sqrt{5}\,a_2/a_0)^2} P_4(\cos\theta) \qquad (14b)$$

for the transition $|{}^1/_2| \xrightarrow{1} |{}^1/_2| \xrightarrow{1} |{}^1/_2|$.

As seen from the plot (Fig. 1b), the sensitivity of the coefficients of the angular distributions to the value of the anisotropy ε is much higher here than in case B.

3. APPARATUS

The measurements were made with the apparatus used by us earlier:[1] the source and detector were mounted at equal distances from the center of the scatterer (200 mm), which constituted a square of 80 × 80 mm, so that the average angle of incidence of the γ rays on the scatterer was equal to the average angle of reflection (focusing geometry). In those cases when it was necessary to make the measurements at low temperatures, the scatterer was secured to the wall of a brass vessel with liquid nitrogen. The thermal insulator was foamed plastic; to prevent frosting of the scatterer, the space in front of it was covered with a thin polyethylene film (see Fig. 2).

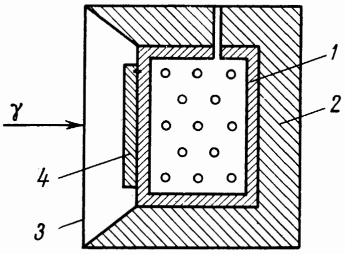

FIG. 2. Cooling system for scatterer: 1—brass vessel filled with liquid nitrogen, 2—foamed-plastic thermal insulation, 3—organic film, 4—scatterer on brass substrate.

In all measurements we used a source in the form of Sn_2O ($f' \approx 0.5$ at $T = 300\,°K$), the emission line of which in our measurements can be regarded as single. The source was rectangular in shape, with height 25 mm and width 12 mm, and its activity was about 0.2 μCi. To tune to resonance with the absorption line in the scatterer, the source was made to execute reciprocating motion with specified speed (up to 5 mm/sec) by means of a miniature mechanical system with a motor. The resonant effect was determined by the difference between the readings corresponding to the forward and backward motion of the source. The measurements were made for scattering angles from 90 to 150°. The corrections for the variation of the solid angle in this range of scattering angles, calculated by the Jaffey formulas,[11] and also determined experimentally, were ~25% (accurate to ~1%). The thickness of the scatterer was chosen in accordance with the condition $C_A = \sigma_0 n f' \geq 10$.[1]

4. RESULTS

A. Measurements of small quadrupole interactions in Mg_2Sn, β-Sn, and SnO_2. In this work, just as before, we determined the attenuation coefficients in the angular-distribution functions from the ratios of the experimental values of the coefficients A_2 (12) of the Legendre polynomials for the investigated substance and for the compound Mg_2Sn, for which, according to investigations of the resonant absorption spectrum,[12] unperturbed correlation should be observed. This method masks possible inaccuracies in the determination of the corrections. Since this makes it obvious that a knowledge of the experimental unperturbed angular-distribution function is necessary here with maximum accuracy, we repeated the measurements with an Mg_2Sn scatterer and obtained results with a lower statistical errors than in [1]. From the approximation of the experimental points (curve 1, Fig. 3) by least squares by means of functions of the type of (12), we obtained

$$A_2(\mathrm{Mg_2Sn}) = 0.251 \pm 0.013,$$

in place of 0.26 ± 0.03 in [1]. The theoretical value for the transition $|{}^1\!/_2| - |{}^3\!/_2| - |{}^1\!/_2|$, corrected for the finite angular resolution, is $A_2 = 0.242$.

Measurements with a scatterer in the form of metallic white tin (β-Sn) were carried out at temperatures 300 and 77° K. The obtained angular distributions are represented by curves 2 and 3 respectively of Fig. 3 and are approximated by the functions

$$W(\theta) = 1 + (0.20 \pm 0.02) P_2(\cos\theta), \quad T = 300^\circ\,\mathrm{K};$$

$$W(\theta) = 1 + (0.17 \pm 0.01) P_2(\cos\theta), \quad T = 77^\circ\,\mathrm{K}.$$

The values of the attenuation coefficients G_2 obtained in this manner and the corresponding values of the quadrupole interactions are listed in Table II. The same table gives these quantities for SnO_2, recalculated from the angular distribution measured in [1] but with the corrected value of $A_2(Mg_2Sn)$ taken into account.

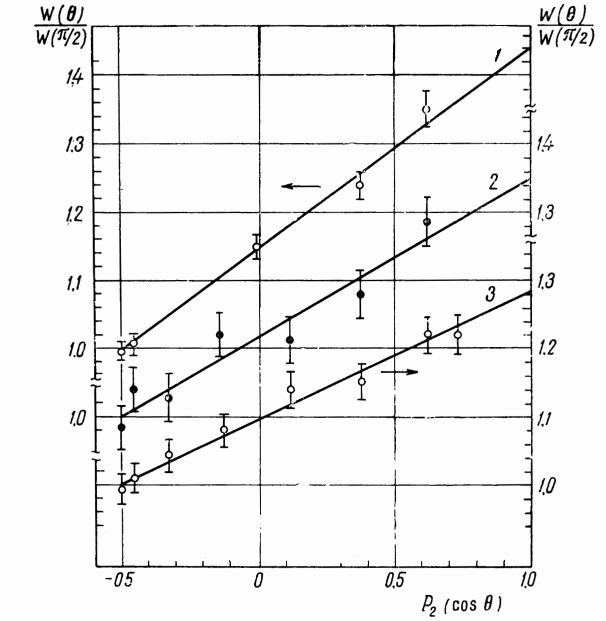

FIG. 3. Angular distributions of the resonant scattering of: 1–Mg_2Sn, 300°K, 2–β-Sn, 300°K, 3–β-Sn, 90°K.

The values of the quadrupole interaction of the Sn^{119} nuclei in β-Sn and SnO_2 lattices were estimated earlier from the shift of the centers of gravity of the resonant-absorption lines in single-crystal samples oriented in various manners,[5, 6] and from the broadening of the absorption lines in polycrystalline samples.[13, 14] In the former case the signs of the quadrupole interactions were determined, but only estimates were obtained for their magnitudes, since the observed shifts were small compared with the experimental line widths. Our results agree within the limits of error with the values of the quadrupole interactions obtained by Mitrofanov et al.[14] from the decomposition of the absorption spectra measured with high apparatus resolution, into unbroadened Lorentz components. Our data, as well as the data of [14], indicate that the quadrupole interaction in β-Sn decreases with rising temperature.

Table II. Coefficients of attenuation of angular distributions and values of the quadrupole interaction for β-Sn and SnO_2. 1) Substance, 2) present work

Substance	T, °K	G_2	E_Q/Γ	Δ, mm/sec	
				present work	[14]
β-Sn	80	0.68±0.07	0.82±0.15	0.25±0.05	0.32±0.02
	300	0.80±0.10	0.58±0.20	0.18±0.06	0.25±0.02
SnO_2	300	0.49±0.05	$1.32^{+0.20}_{-0.14}$	$0.41^{+0.06}_{-0.04}$	0.50±0.04

While the quadrupole splittings for β-Sn and SnO_2 obtained from the absorption spectra and the angular distributions of resonant scattering agree in general within the limits of error, a systematic deviation is noted in the latter case towards lower values (see Table II). This may indicate that the absorption-line broadening in these substances may in part not be of quadrupole origin. (Using formula (10) and the known values of the anisotropy f′ for β-Sn and SnO_2[5, 6] it can be shown that the correction to the coefficients of the angular distributions, which take into account the anisotropy of f′, are in this case negligibly small (~1%) and cannot explain the observed deviation.)

B. Measurements on individual components of the quadrupole doublets of SnO and $(C_4H_9)_2SnO$. As is well known[15, 16] the resonant-absorption spectra in the compounds SnO (tetragonal modification) and $(C_4H_9)_2SnO$ are well-resolved quadrupole doublets with splitting Δ equal to 1.4 and 2.1 mm/sec respectively. This makes it possible to measure the resonant scattering angular distributions corresponding to separate excitation of each of the sublevels.

Measurements with SnO were made at room temperature. The scatterer was prepared by precipitating from a suspension in alcohol of fine powder of black tin oxide, resulting in a dense uniform non-heaping layer over the entire surface of the scatterer. As shown by Boyle et al.[17], the principal axes of the microcrystals become partially oriented in this case in a direction perpendicular to the sample surface. From the resultant asymmetry of the absorption spectrum, the sign of the quadrupole interaction was found in that paper to be positive. Figure 4 shows the absorption spectra for two SnO samples, one of which was prepared by precipitation from the suspension (Fig. 4a) and the other comprising a very fine powder, poured in the measuring cassette (Fig. 4b). In the first case the spectrum is noticeably asymmetrical, and from the degree of asymmetry it is possible, knowing the preferred alignment direction, to determine the relative value of the latter. It turns out to be ~ 10%. The spectrum of the powdered sample was practically symmetrical, indicating the absence of noticeable alignment.

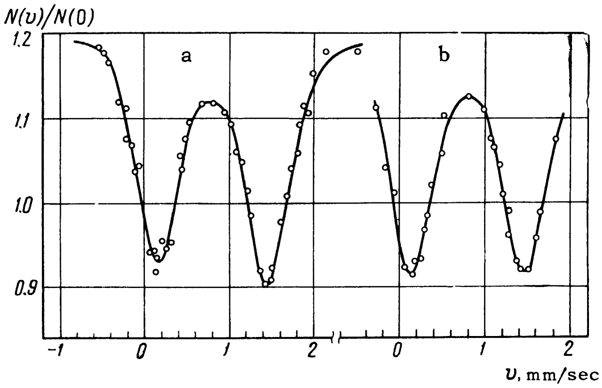

FIG. 4. Absorption spectra for SnO: a—absorber made by precipitation from a suspension; b—absorber in the form of fine powder, N(v)—number of counts as a function of the source speed v.

It is obvious that the partial alignment of the sample should affect the angular distributions. The results of the measurements are shown in Fig. 5, which shows also the theoretical curves for an isotropic sample (the "hard core" is shown dashed) and for a sample with 10% alignment (solid curves). The difference between the calculated curves is especially noticeable for the component of the $|{}^1/_2|-|{}^3/_2|-|{}^1/_2|$ transition (Fig. 5a) (in the case of SnO—the high-energy component), and the experimental points agree satisfactorily with the assumption regarding the indicated degree of alignment of the sample. This fact, and also the symmetry of the spectrum for the powdered sample, indicate that the anisotropy of f′ in this compound is apparently small ($|\epsilon| < 0.15$).

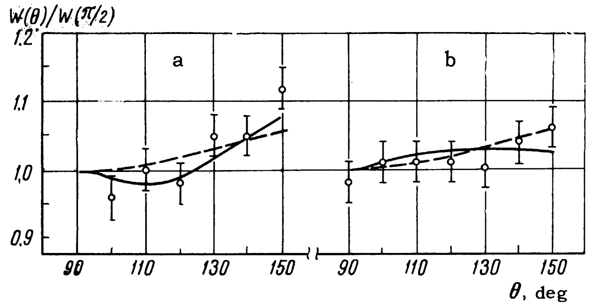

FIG. 5. Angular distributions of resonant scattering for the right-hand (a) and left-hand (b) components of the SnO doublet; T = 300°K. The scatterer was made by precipitation from a suspension. The solid curves are drawn under the assumption of 10% alignment of the microcrystals of the sample in a direction normal to the scatterer plane; the "hard core" is shown dashed.

As already mentioned, in the absorption spectra for organic tin compounds an assymetry of the quadrupole doublets is sometimes observed,[18, 19] one of the possible causes of which may be the anisotropy of the Mossbauer-effect probability.[8]

The compound $(C_4H_9)_2SnO$ (tin dibutyloxide) was chosen from the large group of organic tin compounds characterized by a large quadrupole interaction, because of its chemical stability to variable external conditions (temperature, humidity), this being an important factor for prolonged measurements with periodic heating and cooling of the scatterer.[1]

To obtain an even, uniform, and sufficiently strong scatterer of large thickness ($C_A \approx 10$) from light and friable powder of $(C_4H_9)_2SnO$, possessing in addition a small value of f′ ($f'(77\,°K) \approx 0.2$ summed over both components of the doublet), we constructed it from tablets made by pressing the powder at 150 atm.

Measurements of the angular distributions were made with the scatterer cooled with liquid nitrogen. The results are shown in Fig. 6. It can be seen that the angular distributions for different components of the doublet, unlike those expected for a completely isotropic sample, are markedly different: they have opposite signs, and a negative angular anisotropy is observed for the high-energy component of the doublet. Such a distortion of the angular distributions can be due to two causes (see Sec. 2): 1) the alignment of the microcrystals in the sample, which generally speaking can occur upon compression; 2) the large anisotropy of f′ (see Fig. 1a).

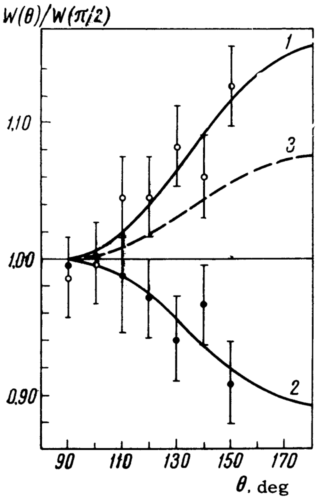

FIG. 6. Angular distributions for the components of the quadrupole doublet of $(C_4H_9)_2SnO$ at T = 80°K. Curve 1–left-hand component, 2–right-hand component (see Fig. 7). The solid curves are drawn through the experimental points by least squares, and the dashed curve is the "hard core."

Both factors should affect also the form of the resonant-absorption spectra. These spectra were measured by us with samples of two types: prepared from finely crushed powder, or pressed into a tablet at 150 atm. The sample thickness was $C_A \approx 1$. Both spectra (Fig. 7) are markedly asymmetrical, the degree of asymmetry being the same for both within the limits of error and amounting to ~ 10% (as determined from the areas under the lines).

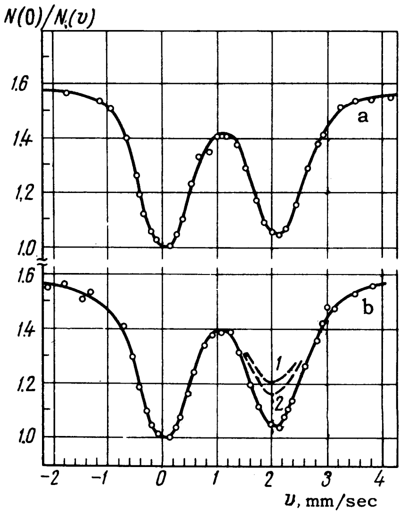

FIG. 7. Absorption spectra of $(C_4H_9)_2SnO$, T = 80°K: a–powdered absorber, b–pressed absorber. In spectrum b the dashed curves are the calculated intensities of the right-hand component under the alignment assumed in the text for the sample.

Calculation by formulas (7a) and (7b) of Sec. 2 shows that to attribute the observed negative anisotropy of the angular distribution for one of the components to the first of the aforementioned causes it is necessary to assume either a 20% degree of alignment in a direction perpendicular to the plane of the sample (in which case this is the component with $|m_b| = 1/2$), or 40% in the scatterer plane (in which case this is the component with $|m_b| = 3/2$). The dashed curves on the absorption spectrum show the intensities expected in these cases for the right-hand (high-energy) component relative to the left-hand component. So large a change in the relative intensities of the component could not go unnoticed, and comparison of the spectra of Figs. 7a and 7b shows that the sample apparently remains isotropic after compression.

On the other hand, the entire aggregate of the observed facts—the asymmetry of the absorption spectrum

$$I_{\text{right}} / I_{\text{left}} = 0.90 \pm 0.06$$

and the angular distributions

$$W_{\text{left}}(\theta) = 1 + (0.10 \pm 0.03) P_2(\cos\theta),$$
$$W_{\text{right}}(\theta) = 1 - (0.06 \pm 0.03) P_2(\cos\theta)$$

have a natural explanation if it is assumed that the

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probability of the effect f′ in crystals of $(C_4H_9)_2SnO$ is strongly anisotropic. Simultaneous solution of (11a) and (11b) with respect to a_2/a_0, using the indicated experimental values of the coefficients of the Legendre polynomials in the angular distributions, is possible at values $\epsilon < -0.6$. The left-hand and right-hand components of the doublet in the absorption spectrum correspond in this case to $|m_b| = 1/2$ and $|m_b| = 3/2$ respectively, i.e., $eQV_{zz} > 0$. However, the accuracy of the present measurements does not exclude completely the possibility of $\epsilon \sim 0.5$, (in which case $eQV_{zz} < 0$). At any rate, it can be concluded that the probabilities of recoilless absorption in directions parallel and perpendicular to the axis of the molecular crystal $(C_4H_9)_2SnO$, differ by at least a factor of 3. So strongly pronounced an anisotropy should be accompanied by a considerable anisotropy of the elastic properties.

Strictly speaking, comparison of the experimental angular distributions with formulas (11a) and (11b) is valid if the molecular crystals $(C_4H_9)_2SnO$ are axially-symmetrical and the angular dependence of f′ can be represented in the form (8). This question calls for a special investigation.

5. CONCLUSION

From the results of the present paper it follows that measurements of the angular distributions of the Mossbauer resonant scattering can be used not only to determine the value of the quadrupole interaction (it should be noted that such a determination is direct and does not depend on such factors as inaccurate knowledge of the value of f′, which can introduce an uncertainty in the value of Δ determined from the absorption spectrum when $\Delta \sim \Gamma$), but also to disclose effects connected with the dynamics of the crystal lattice, particularly the anisotropy of the Mossbauer effect. It must be particularly emphasized that in principle one can determine simultaneously the magnitude and the sign of the anisotropy and the sign of the quadrupole interaction without using single-crystal samples, something impossible with absorption-spectrum measurements. Since single-crystal samples are in most cases rather difficult and sometimes impossible to produce, the number of objects that can be investigated through the use of this method is greatly increased.

At large values of the anisotropy of f′ ($|\epsilon| > 0.3$), the indicated quantities can be relatively easily determined at a moderate measurement accuracy. In order to proceed to smaller values, it is necessary to increase the accuracy of the determined angular-distribution coefficients to ~5% at $A_2 \approx 0.05$ (in the case of Sn^{119}), which is not difficult. As already noted (item C of Sec. 2), when a resonant detector is used the angular distributions of the resonant scattering are more sensitive to the degree of anisotropy of f′. Therefore, in spite of additional experimental difficulties (loss of intensity at small values of f′, some complication of the experimental setup), such measurements may be useful for this purpose.

The increase in measurement accuracy should also be accompanied by an increase in the computation accuracy: it is necessary to take into account such factors as the non-axiality of the electric field gradient, the non-coincidence of the gradient and crystal axes, possible relaxation processes which may particularly strongly affect the intermediate state of the nucleus, etc.

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