

RESONANCE ABSORPTION OF 23.8-keV GAMMA QUANTA BY  $\text{Sn}^{119}$  NUCLEI IN CRYSTALS

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Submitted to JETP editor April 27, 1961

J. Exptl. Theoret. Phys. (U.S.S.R.) **41**, 1347-1358 (November, 1961)

Using a source of  $\text{Sn}^{119\text{m}}$  in  $\text{SnO}_2$ , we have studied the resonance absorption of 23.8-keV  $\gamma$  quanta by  $\text{Sn}^{119}$  nuclei in crystals of  $\text{SnO}_2$ ,  $\text{SnO}$ ,  $\beta\text{-Sn}$  and  $\text{SnNb}_3$ . Resonance absorption spectra were obtained for different absorber thicknesses at liquid nitrogen temperature, and at solid  $\text{CO}_2$  temperature for the  $\beta\text{-Sn}$  crystal. For each of the crystals we determined the probability of absorption without energy loss to recoil, and the magnitude of the isomer shift relative to the energy of the  $\gamma$  transition in the  $\text{SnO}_2$  crystal. For crystals of  $\text{SnO}$  and  $\beta\text{-Sn}$ , we observed quadrupole splittings of the absorption line, which were equal (at liquid nitrogen temperature) to  $(12.5 \pm 1.5) \times 10^{-8}$  and  $(11.0 \pm 1.5) \times 10^{-8}$  eV, respectively. For the  $\beta\text{-Sn}$  crystal, the quadrupole splitting has a strong temperature dependence.

## 1. INTRODUCTION

AS was first shown by Mössbauer,<sup>[1]</sup> nuclei contained in crystal lattices have a certain probability for radiating (or absorbing)  $\gamma$  quanta without loss of energy to nuclear recoil. The quantum state of the crystal does not change and the recoil momentum is transferred to the crystal as a whole; since the mass of the crystal can be regarded as infinitely large, there is no loss of energy of the quantum as a result of the recoil. The probability for such a process, calculated in the Debye approximation using Lamb's theory,<sup>[2]</sup> coincides with the Debye-Waller factor which appears in coherent x-ray scattering. This probability increases with decreasing recoil energy of the free atom  $R = E_0^2/2Mc^2$  (where  $E_0$  is the energy of the  $\gamma$  transition and  $M$  is the mass of the atom), with decreasing temperature of the crystal and increase of its Debye temperature. Thus we may expect a sizable probability of the Mössbauer effect for low energy  $\gamma$  transitions ( $E_0 \leq 100$  keV); in addition in most cases it is necessary to resort to cooling the source and absorber.

The experimental data which have been obtained and the theoretical computations, which have been made by Kagan,<sup>[3]</sup> show that the Debye temperature is generally not the determining factor of the probability for this process, except possibly for the case of monatomic crystals. The probability for recoilless emission or absorption of  $\gamma$  quanta depends essentially on the specific form of the vibrational spectrum of the crystal lattice. The fundamental theory of the Mössbauer effect and the

relation of its probability to crystal properties is treated in the papers of Visscher,<sup>[4]</sup> Lipkin,<sup>[5]</sup> and Shapiro.<sup>[6]</sup>

The presence in the emission and absorption spectra of lines having natural width and not subject to any Doppler broadening or shift due to recoil provides favorable conditions for observing the resonance absorption of  $\gamma$  quanta in crystals. By giving the source a velocity  $v$  relative to the absorber, we can artificially produce a Doppler shift of the absorption line by an amount  $\Delta E = E_0 v/c$ . By observing the resulting change in the effective cross section for the resonance absorption, we can investigate various effects associated with very small changes in the energy of the  $\gamma$  transition. The required velocities are equal to a few millimeters or centimeters per second. Experiments carried out recently using the Mössbauer effect are discussed in the published summaries.<sup>[7]</sup>

In the present work, we investigate the spectra of resonance absorption of 23.8-keV  $\gamma$  quanta by  $\text{Sn}^{119}$  nuclei contained in the crystal lattices of various tin compounds. Some preliminary results of these experiments have been published previously.<sup>[8,9]</sup>

## 2. CALCULATION OF MAGNITUDE OF RESONANCE ABSORPTION

In studying the dependence of the resonance absorption on the relative velocity  $v$  of source and absorber, the quantity which is measured experimentally is

$$\varepsilon(v) = [N(\infty) - N(v)]/N(\infty), \quad (1)$$

where  $N(v)$  is the counting rate for  $\gamma$  quanta transmitted through the absorber when the velocity is  $v$ ;  $N(\infty)$  is the counting rate at high velocity, where the resonance absorption is absent. In the future, for brevity, we shall call the quantity  $\varepsilon(v)$  the magnitude of the effect. It is not hard to show that the expression (1) for the magnitude of the effect can be written in the form

$$\varepsilon(v) = \kappa f \left( 1 - \int_0^{\infty} e^{-\sigma(E)n} W_e(E) dE \right), \quad (2)$$

where  $\sigma(E)$  is the effective cross section for resonance absorption of  $\gamma$  quanta of energy  $E$ ,  $W_e(E)$  is the distribution function for the emission spectrum,  $n$  is the number of atoms of the particular isotope per  $\text{cm}^2$  of the absorber,  $f$  is the probability of recoilless radiation of the  $\gamma$  quanta,  $\kappa$  is a parameter which determines the relative contribution of the particular  $\gamma$  quanta to the total counting rate. The parameter  $\kappa$  takes account of the presence of background, i.e., the recording by the counter of  $\gamma$  radiation whose resonance absorption is not under investigation.

The effective cross section is given by the formula

$$\sigma(E) = \frac{\Gamma^2}{4} \sigma_0 \frac{f'}{(E - E_0)^2 + \Gamma^2/4}, \quad (3)$$

where  $\Gamma$  is the total width of the level,  $f'$  is the probability of recoilless absorption of the  $\gamma$  quanta, and

$$\sigma_0 = \frac{2I_1 + 1}{2I_0 + 1} \frac{\lambda^2}{2\pi} \frac{\Gamma_\gamma}{\Gamma}.$$

(Here  $I_0$  and  $I_1$  are the spins of the ground and excited states,  $\lambda$  is the wave length of the  $\gamma$  radiation,  $\Gamma_\gamma$  is the radiative width; for the first excited state,  $\Gamma_\gamma/\Gamma = 1/(1 + \alpha)$  where  $\alpha$  is the total internal conversion coefficient.)

In the following we shall, for simplicity, consider the case where the source and absorber are chemically identical and are at the same temperature i.e., the case where there is an exact overlap of the emission and absorption lines for  $v = 0$ . This does not cause any loss of generality, since the presence of a constant line shift  $\delta$  can be taken into account by simply changing the velocity  $v$  to  $v + \delta$ .

If the source does not contain the nuclei whose resonance absorption is being studied (or if the source is sufficiently thin) the emission spectrum has the form

$$W_e(E) = \frac{\Gamma}{2\pi} \frac{1}{(E + E_0/c - E_0)^2 + \Gamma^2/4}, \quad (4)$$

and the magnitude of the effect can be written in the following form:

$$\varepsilon(v) = \kappa f \left( 1 - \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\exp[-C/(1+x^2)]}{1+(x+y)^2} dx \right),$$

$$x = \frac{E - E_0}{\Gamma/2}, \quad y = \frac{E_0 v/c}{\Gamma/2}, \quad C = \sigma_0 f' n. \quad (5)$$

The integral in (5) can in general be computed only by numerical integration. An exception is the case of  $v = 0$ , where the expression (5) can be brought to the form (cf., for example, [10]):

$$\varepsilon(0) = \kappa f [1 - I_0(C/2)e^{-C/2}], \quad (6)$$

where  $I_0(x) = J_0(ix)$  is the Bessel function of zero order and imaginary argument.

Expression (6) can be used to determine the value of  $f'$ , if we determine experimentally the dependence of the magnitude of the effect on absorber thickness. Formula (6) contains two unknown parameters ( $f'$  and  $\kappa f$ ), so it is convenient to make a comparison with experiment of the ratio  $\varepsilon(0, n)/\varepsilon(0, n_0)$ , where  $n_0$  is some fixed value of  $n$ .

Formula (5) enables us to compute the shape of the experimentally measured absorption spectrum. In particular we can find the width of the line in the resonance absorption spectrum as a function of the absorber thickness. For sufficiently thin absorbers ( $C \ll 1$ ) the line width at half maximum is equal to  $2\Gamma$ ; with increasing absorber thickness the line width increases because of the effect of saturation. The natural width of the level can be found by extrapolating to zero thickness or by making corrections to the observed width in accordance with formula (5); in the latter case a prior determination of  $f'$  is required.

If the source contains nuclei of the isotope whose resonance absorption is being investigated, formulas (5) and (6) are not exact, since the emission spectrum will be distorted because of self-absorption and cannot be represented by formula (4). It is easy to show that in this case the distribution function for the emission spectrum will have the form (for  $v = 0$ ):

$$W_e(E) = \text{const} \cdot \frac{1 - \exp\left[-\frac{\mu d_0 + C + \mu d_0 x^2}{1 + x^2}\right]}{\mu d_0 + C + \mu d_0 x^2}, \quad (7)$$

where  $C = \sigma_0 f' n$ ,  $d_0$  is the source thickness and  $\mu$  is the coefficient of nonresonant absorption. We note that the function (7) practically coincides with expression (4) when  $v = 0$ , if we simply replace the natural width  $\Gamma$  by a width  $\Gamma_{\text{eff}}$ , which is larger than  $\Gamma$ . So for practical computations

formula (7) can be written in the form

$$W_e(E) = b \frac{\Gamma}{2\pi} \frac{1}{b^2(E - E_0)^2 + \Gamma^2/4}, \quad (8)$$

where  $b = \Gamma/\Gamma_{\text{eff}}$ . The parameter  $b$  can be calculated for different values of  $f'$  by using formula (7).

If the nucleus has electric or magnetic moments (in its ground or excited states), then as a result of interaction with external or crystalline fields the emitted line, the absorption line or both may be split into a number of components. In this case the results obtained above cannot be used directly since every splitting causes a change in the form of the functions  $\sigma(E)$  and  $W_e(E)$ .

The simplest case is that in which the emission line is split into a number of components with relative intensities  $J_i$  ( $\sum J_i = 1$ ), while the absorption line is unsplit. From formula (2) it immediately follows that in this case the magnitude of the effect can be represented as a sum of terms like (2), each of which enters with weight  $J_i$ . The presence of a shift between the  $i$ -th component of the emission line and the absorption line must be taken into account by an appropriate choice of the form of the function  $W_e(E)$ . It is easy to see that for the case of such a splitting the magnitude of the effect can be written in the form

$$\epsilon(v) = \sum_i J_i \epsilon(\delta_i), \quad (9)$$

where  $\delta_i$  is the shift between the  $i$ -th component of the emission spectrum and the absorption line, for a given relative velocity of source and absorber.

The computation is more difficult for the case where the absorption line is split. Then the function  $\sigma(E)$  splits into a sum of terms:  $\sigma(E) = \sum_i J_i \sigma(E + \delta_i)$ , where  $J_i$  is the relative intensity of the  $i$ -th component in the absorption spectrum, and  $\delta_i$  is the shift between the emission line and the  $i$ -th component of the absorption line. Since the function  $\sigma(E)$  is in the exponent of the formula (2), in this case the expression for the magnitude of the effect cannot be written as a sum of terms, as we did above. The computation of the quantity  $\epsilon(v)$  can be done only by numerical integration.

Let us consider in more detail the case of quadrupole splitting of the absorption line for the case of  $\text{Sn}^{119}$ . The interaction of the nuclear quadrupole moment with the electric field gradient  $q_{ij} = \partial^2 V / \partial x_i \partial x_j$  is described by the Hamiltonian

$$\mathcal{H} = \frac{eQ}{2I(2I-1)} \sum_{i,j} q_{ij} I_i I_j,$$

where  $Q$  is the nuclear quadrupole moment and

$I$  is the nuclear spin operator. As a result of such an interaction, the excited state of the  $\text{Sn}^{119}$  nucleus (which has spin  $3/2$ ) splits into two sublevels, which are degenerate in the sign of the spin projection; their separation is

$$\Delta = \frac{1}{2} e^2 Q q_{zz} \left(1 + \frac{1}{3} \eta^2\right)^{1/2}, \quad (10)$$

where the asymmetry parameter  $\eta = (q_{xx} - q_{yy})/q_{zz}$ .

The ground state of  $\text{Sn}^{119}$  (with spin  $1/2$ ) is not split by quadrupole interaction. Consequently the absorption spectrum should consist of two lines, of equal intensity, whose separation is given by formula (10). Let us find the expression for the magnitude of the effect for the case where, at a given velocity  $v_0$ , the unsplit emission line exactly overlaps one of the two components of the absorption line. Using formulas (2) and (5) one can easily show that in this case the magnitude of the effect is

$$\epsilon(v_0) = \kappa f \left(1 - \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\exp\left\{-\frac{C}{2} \left(\frac{1}{1+x^2} + \frac{1}{1+(x+y)^2}\right)\right\}}{1+x^2} dx\right), \quad (11)$$

where  $x$  has the same meaning as in (5), and  $y = \Delta(\Gamma/2)^{-1}$ . The integral in (11) was calculated by numerical integration for different values of the parameters  $C$  and  $\Delta$ . The results were used for determining the values of  $f'$  for cases of quadrupole splitting of the absorption line.

### 3. DESCRIPTION OF THE EXPERIMENT

Measurements of the dependence of the resonance absorption effect on the relative velocity of source and absorber (the resonance absorption spectrum) were made with an apparatus whose principle of operation was described briefly earlier.<sup>[8]</sup> An overall view of the arrangement (omitting the electronics) is shown in Fig. 1. The absorber 9 moved relative to the fixed source 5, where the absorber velocity changed linearly with time between given limits (from  $-v_0$  to  $+v_0$ , where  $v_0$  is the maximum velocity). The absorber velocity was produced by rotation of the cam 2, whose profile was cut to correspond to a linear variation of velocity. The rate of rotation of the cam (and consequently the limits of variation of the absorber velocity) could be changed by a system of gears and pulleys. The cam set in motion a rod to which the frame-holder 4 of the absorber was attached.

The source was at room temperature in all the measurements. In many of the measurements the absorbers were cooled with liquid nitrogen or dry ice; in these cases the absorbers were placed in a

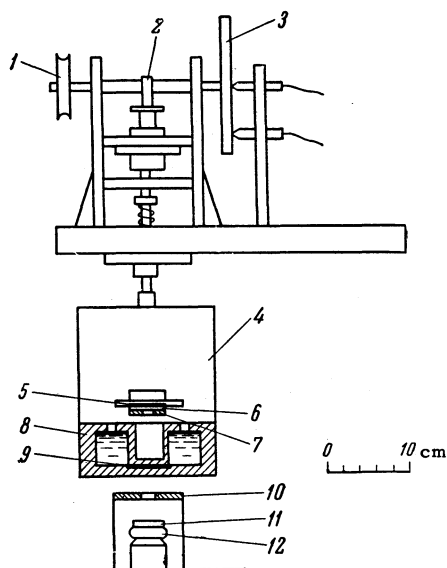


FIG. 1. Schematic diagram of apparatus for measuring resonance absorption spectra: 1 – pulley, 2 – cam, 3 – contact disc for amplitude modulation circuit, 4 – absorber holder, 5 – source, 6 – palladium filter, 7 – lead collimator, 8 – low-temperature styrofoam vessel, 9 – absorber, 10 – lead collimator, 11 – NaI(Tl) crystal, 12 – photomultiplier.

low-temperature vessel 8 of styrofoam. The construction of the container was such that a  $\gamma$  quantum on its path to the counter encountered no liquid nitrogen but only styrofoam, which absorbs the 24 keV  $\gamma$  quanta weakly. The x rays of tin are practically completely absorbed by the 0.6 mm thick characteristic filter 6 of palladium.

Gamma quanta passing through the absorber were recorded by a NaI(Tl) crystal 1.5 mm thick and a FEU-11 photomultiplier. The amplified pulses entered a single-channel pulse analyzer, whose window was set on the 24 keV photopeak. Separate recording of counts of  $\gamma$  quanta for different absorber velocities was accomplished by linear (in time) amplitude modulation of the pulses. The pulses from the output of the single-channel analyzer entered a gating circuit at whose other input a modulating voltage was applied which increased linearly in time in synchronism with the change in velocity of the absorber. The amplitude-modulated pulses entered a 100-channel AI-100 pulse analyzer. The period of variation of the modulation voltage was exactly equal to the period of rotation of the cam; thus there was a one-to-one correspondence between absorber velocity and amplitude of the output pulse, which enabled us to measure the whole absorption spectrum simultaneously, i.e., the dependence of the resonance absorption effect on velocity over the whole selected range of velocities. In the present work measurements were made for three different values of the maximum velocity: 2.5, 5, and 6.5 mm/sec.

A source in the form of  $\text{SnO}_2$  was prepared from metallic tin, enriched to 92% of  $\text{Sn}^{118}$  and then irradiated with thermal neutrons in a reactor.

The powder of irradiated tin was then oxidized by heating to form  $\text{SnO}_2$ ; at the beginning the  $\text{SnO}_2$  is formed mainly in an amorphous state. After a long period of baking (ten days) at a temperature  $\sim 700^\circ\text{C}$  a transformation of the  $\text{SnO}_2$  to the metallic state occurs. The source thickness was 6  $\text{mg}/\text{cm}^2$ .

As was already mentioned earlier, for the  $\text{SnO}_2$  crystal the probability of recoilless emission of  $\gamma$  quanta is already reasonably large at room temperature. There is no quadrupole splitting of the emission line for the  $\text{SnO}_2$  crystal. Thus the source of  $\text{Sn}^{119\text{m}}$  in the form of tin dioxide is convenient for the investigation of the resonance absorption of 23.8-keV  $\gamma$  quanta by  $\text{Sn}^{119}$  and has definite advantages over, for example, the source of white metallic tin previously used by us. [8,9]

Absorbers of white metallic tin were prepared by rolling tin foil or by depositing tin on an organic backing in vacuum. Absorbers of  $\text{SnO}_2$ ,  $\text{SnO}$  and  $\text{SnNb}_3$  were prepared by precipitating powders on a thin aluminum foil.

#### 4. RESULTS OF EXPERIMENT

In all the measurements, the  $\text{Sn}^{119\text{m}}\text{O}_2$  source was at room temperature, while the absorbers were either at room temperature or at liquid nitrogen temperature or (for the white tin absorbers) at dry ice temperature.

$\text{SnO}_2$ . The measurements of the resonance absorption spectrum for a  $\text{SnO}_2$  polycrystal were done with absorbers of seven different thicknesses in the range from 4.2 to 37  $\text{mg}/\text{cm}^2$ . In Fig. 2 we show one of these spectra, for an absorber thickness of 9.4  $\text{mg}/\text{cm}^2$  at room temperature. For each of the measured spectra we determined the

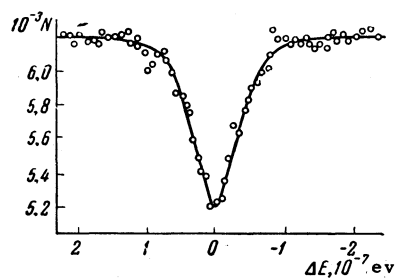


FIG. 2. Resonance absorption spectrum for an  $\text{SnO}_2$  absorber. The abscissa is  $\Delta E = E_0 v/c$  (where  $E_0 = 23.8$  keV and  $v$  is the absorber velocity). Positive values of  $\Delta E$  correspond to motion of the absorber toward the source. The ordinates give the number of quanta passing through the absorber, which is proportional to the number  $N$  of quanta recorded.

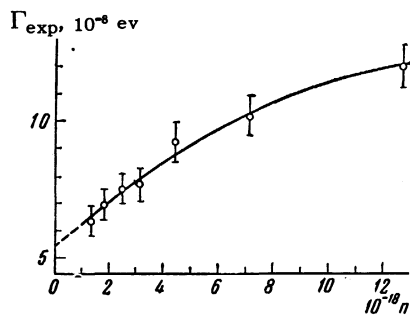


FIG. 3. Dependence of width of absorption line  $\Gamma_{\text{exp}}$  on absorber thickness. The abscissa gives the number of atoms containing the  $\text{Sn}^{119}$  isotope per  $\text{cm}^2$  of absorber.

width of the absorption at half height  $\Gamma_{\text{exp}}$  and the maximum value of the resonance absorption effect  $\epsilon(0)$ .

The dependence of the line width of the absorption spectrum on absorber thickness is shown in Fig. 3. Extrapolation to zero absorber thickness gives the value  $(5.5 \pm 0.5) \times 10^{-8}$  eV. This is equal to the sum of the natural width  $\Gamma$  and the width of the line in the emission spectrum. A computation shows that for the source which we used (containing 2.3% of  $\text{Sn}^{119}$ ) the broadening of the line in the emission spectrum is only 12%. Thus we get for the natural width of the 23.8 keV excited state of  $\text{Sn}^{119}$  the value

$$\Gamma = (2,60 \pm 0,25) \cdot 10^{-8} \text{ eV},$$

which is in good agreement with the value found earlier by a delayed coincidence method.<sup>[11]</sup>

The probability for recoilless absorption of the  $\gamma$  quanta,  $f'$ , was determined for the  $\text{SnO}_2$  crystal from the dependence of  $\epsilon(0)$  on absorber thickness. From formula (6), for different values of  $f'$  we can calculate the dependence on  $n$  of the ratio  $\epsilon(0, n)/\epsilon(0, n_0)$ . To improve the accuracy and reliability of the results, we took for  $n_0$  in turn the thicknesses of all the seven absorbers which we used. In Fig. 4, the dependence is shown for  $n_0 = 1.45 \times 10^{18}$  ( $4.2 \text{ mg/cm}^2$ ).

The average value of  $f'$  for the  $\text{SnO}_2$  crystal at room temperature was found to be  $0.7 \pm 0.1$ . With the absorber cooled to liquid nitrogen temperature, the value of  $f'$  is close to unity. Knowing the value of  $f'$  enables us to determine the parameter  $\kappa f$  [cf. formula (6)] for the source. It was equal to  $0.25 \pm 0.02$ . The parameter  $\kappa f$  was much less than unity, mainly because the source radiation contained x rays of indium produced from decay of the long-lived isotope  $\text{Sn}^{113}$ . Since this parameter is practically independent of the form of the absorber, this value can now be used for determining  $f'$  for the other crystals.

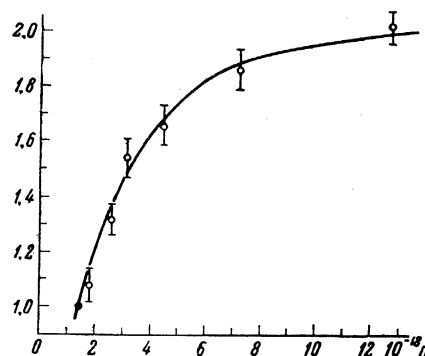


FIG. 4. Dependence of maximum value of resonance absorption effect  $\epsilon(0)$  on thickness of  $\text{SnO}_2$  absorber. The abscissa is the number of atoms containing the  $\text{Sn}^{119}$  isotope per  $\text{cm}^2$  of absorber. The ordinate is  $\epsilon(0, n)/\epsilon(0, n_0)$ , where  $n_0 = 1.45 \times 10^{18}$ . The solid curve was calculated theoretically for  $f' = 0.7$ .

SnO. Resonance absorption spectra for a  $\text{SnO}$  polycrystal were measured with absorbers of various thicknesses in the range from  $4.8$  to  $27 \text{ mg/cm}^2$ , at liquid nitrogen temperature and at room temperature. Two of these spectra are shown in Fig. 5. Both spectra have a doublet structure, corresponding to a splitting of the absorption line in the  $\text{SnO}$  crystal into two components. Such a splitting can be interpreted as the result of the interaction of the quadrupole moment of the  $\text{Sn}^{119}$  nucleus in its excited state with the electric field gradient in the  $\text{SnO}$  crystal. The center of the absorption line is shifted toward positive energies relative to the emission line.

It is apparent that changes of temperature have little effect on the appearance of the absorption spectrum (in contrast to the case of white tin, which will be considered later). The value of the shift of the absorption line in the  $\text{SnO}$  crystal relative to the energy of the emission line in an  $\text{SnO}_2$

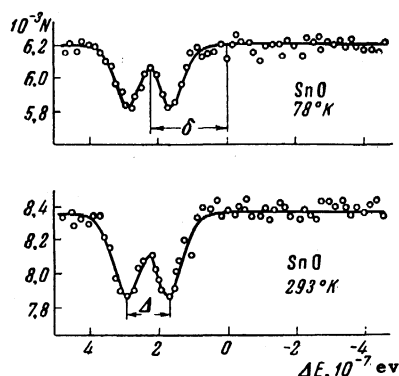


FIG. 5. Absorption spectrum for  $\text{SnO}$  crystal. The upper figure is for a  $4.8 \text{ mg/cm}^2$  absorber at liquid nitrogen temperature. The lower is for a  $15 \text{ mg/cm}^2$  absorber at room temperature.

crystal is equal to  $(22 \pm 2) \times 10^{-8}$  at  $78^\circ\text{K}$  and  $(21.5 \pm 1.5) \times 10^{-8}$  eV at  $293^\circ\text{K}$ . The values of the quadrupole splitting  $\Delta$  are equal to  $(12.5 \pm 1.5) \times 10^{-8}$  eV at  $78^\circ\text{K}$  and  $(11.0 \pm 1.5) \times 10^{-8}$  eV at  $293^\circ\text{K}$ .

The probability  $f'$  for recoilless absorption of  $\gamma$  quanta by the SnO crystal was determined from the dependence of the magnitude of the effect on absorber thickness. The experimental values of the effect were compared with those computed for different values of  $f'$  according to formula (11). We found a value of  $0.47 \pm 0.07$  at liquid nitrogen temperature and  $0.15 \pm 0.05$  at room temperature.

**$\beta$ -Sn.** The resonance absorption of 23.8 keV  $\gamma$  quanta by  $\text{Sn}^{119}$  nuclei in a polycrystal of white tin ( $\beta$ -Sn) was first observed in the work of Lyubimov and Alikhanov<sup>[12]</sup> and of Barloutaud et al.<sup>[13]</sup> In our previous work,<sup>[8,9]</sup> in which we used a source of  $\text{Sn}^{119\text{m}}$  in the form of a  $\beta$ -Sn polycrystal, we showed that at liquid nitrogen temperature the absorption line (or the emission line) in a  $\beta$ -Sn crystal is split by the quadrupole interaction into two components whose separation  $\Delta$  is equal to  $(11.0 \pm 1.5) \times 10^{-8}$  eV.

The observation of the quadrupole splitting with source and absorber in the form of polycrystals of  $\beta$ -Sn presents considerable difficulties. The structure of the absorption spectrum is well resolved only if one uses a source and absorber which are thin (in the  $\text{Sn}^{119}$  isotope). If this is not the case, the broadening of the emission line in the source because of self-absorption and of the absorption line because of saturation prevents a satisfactory separation of the side maxima from the central absorption maximum, which has an amplitude twice that of the side maxima.

The effect of absorber thickness on the shape of the absorption spectrum for this case is graphically illustrated by Fig. 6, in which are shown absorption spectra in a  $\beta$ -Sn crystal at liquid nitrogen temperature, obtained with a source of white metallic tin, which was also at liquid nitrogen temperature. Despite the fact that the source was very thin ( $9 \text{ mg/cm}^2$ , content of  $\text{Sn}^{119}$  isotope equal to 2.3%), for an absorber thickness of  $31.3 \text{ mg/cm}^2$  the side maxima are poorly resolved from the central one. The considerable broadening of the line may explain the result of Picou et al.,<sup>[14]</sup> who used relatively thick sources and absorbers and did not observe any quadrupole splitting in  $\beta$ -Sn at liquid nitrogen temperatures.

The use of an  $\text{Sn}^{119\text{m}}$  source in a polycrystal of  $\text{SnO}_2$ , in which there is no quadrupole splitting of the line, considerably simplifies the problem of investigating the hyperfine structure of the ab-

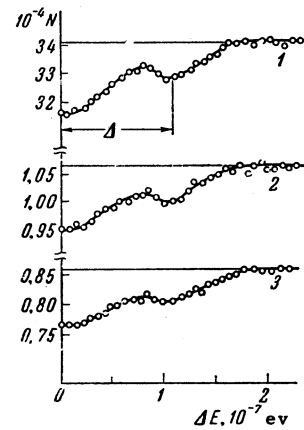


FIG. 6. Resonance absorption spectrum for a  $\beta$ -Sn crystal, obtained with a source of  $\text{Sn}^{119\text{m}}$  in the form of white metallic tin. The absorber thicknesses were: 1 –  $5 \text{ mg/cm}^2$ , 2 –  $14.4 \text{ mg/cm}^2$ , 3 –  $31.3 \text{ mg/cm}^2$ . The spectra are symmetric about zero velocity, so the figure shows only the half of the spectrum for positive velocities.

sorption line in the  $\beta$ -Sn crystal, though even in this case the source and absorber must be sufficiently thin.

The measurements of the resonance absorption spectrum for a  $\beta$ -Sn polycrystal were done with absorbers of varying thickness in the range from 5 to  $50 \text{ mg/cm}^2$  of natural tin, at liquid nitrogen temperature, dry ice temperature and room temperature. The main results of these measurements were discussed in our previous paper,<sup>[15]</sup> where we gave the resonance absorption spectra. It was shown that the quadrupole splitting  $\Delta$  in the  $\beta$ -Sn crystal depends strongly on temperature. We found the following values for  $\Delta$ :  $(11.1 \pm 1.5) \times 10^{-8}$  eV at liquid nitrogen temperature,  $(8.0 \pm 1.6) \times 10^{-8}$  eV at dry-ice temperature and  $(4.6 \pm 1.5) \times 10^{-8}$  eV at room temperature.

Thus, with increasing temperature the value of the quadrupole interaction decreases, and at room temperature the splitting  $\Delta$  does not exceed the line width in the absorption spectrum ( $5 \times 10^{-8}$  eV) even for a thin source and absorber. In this case one can only judge the quadrupole interaction from the considerable broadening of the line in the absorption spectrum. This fact may explain the results of Boyle et al.,<sup>[16]</sup> who observed no splitting of the line from  $\beta$ -Sn with the absorber at room temperature. In their work they used a thick source for which the width of the emission line was twice the natural width; under such conditions of measurement, the quadrupole splitting of the line could not be seen.

The probability of recoilless absorption of  $\gamma$  quanta,  $f'$ , was determined for the  $\beta$ -Sn crystal by the method discussed above for the case of the

SnO crystal. For  $f'$  at liquid nitrogen temperature we obtained the value  $0.4 \pm 0.1$ , which is in good agreement with the value of 0.36, found by Picou et al.<sup>[14]</sup> The determination of  $f'$  at room temperature for the case of  $\beta$ -Sn is difficult, because at this temperature the hyperfine structure components are not resolved and the magnitude of the quadrupole interaction is not determined sufficiently accurately. From an analysis of the line shape in the absorption spectrum and of the dependence of the magnitude of the effect on absorber thickness, we found for  $f'$  at room temperature a value of the order of 0.1. Picou et al.<sup>[14]</sup> found a value  $0.04 \pm 0.01$  for  $f'$  at room temperature, but they did not take into account the change in the quadrupole splitting of the absorption line with temperature.

**SnNb<sub>3</sub>.** We have shown previously<sup>[8]</sup> that there is no quadrupole splitting of the absorption line in the SnNb<sub>3</sub> crystal and that at liquid nitrogen temperature the energy of the  $\gamma$  transition in the SnNb<sub>3</sub> crystal is close to the energy in a crystal of white tin. Resonance absorption spectra obtained with the absorber temperature equal to that of liquid nitrogen and room temperature, respectively, are shown in Fig. 7. A striking feature is the considerable reduction in the energy of the  $\gamma$  transition with increasing temperature of the crystal. The shift in energy of the  $\gamma$  transition in the SnNb<sub>3</sub> crystal is equal to  $+(15 \pm 2) \times 10^{-8}$  eV at liquid nitrogen temperature and  $+(11 \pm 2) \times 10^{-8}$  eV at room temperature. The values of  $f'$  were determined from the dependence of the magnitude of the effect on absorber thickness, and were equal to  $0.3 \pm 0.1$  at liquid nitrogen temperature and  $0.06 \pm 0.02$  at room temperature.

## 5. DISCUSSION OF RESULTS

The results of our measurements of the resonance absorption spectra of the 23.8 keV  $\gamma$  quanta

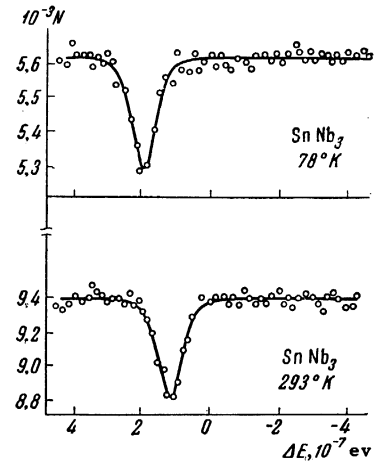


FIG. 7. Absorption spectra of SnNb<sub>3</sub> crystal. Upper figure – 26 mg/cm<sup>2</sup> absorber at liquid nitrogen temperature; lower figure – 62 mg/cm<sup>2</sup> absorber at room temperature.

by Sn<sup>119</sup> nuclei in crystals of SnO<sub>2</sub>, SnO,  $\beta$ -Sn and SnNb<sub>3</sub> are given in the table.

The values found for the probability of absorption of  $\gamma$  quanta without loss of energy to recoil vary over quite a wide range for the various crystals. It is not possible at present to make a detailed comparison of the experimentally determined values of  $f'$  with the theoretical computations, since such computations can be carried out only if one knows the phonon spectrum of a real crystal. Computations on the Debye approximation are certainly only a rough approximation; the Debye temperature cannot in general serve as a parameter to determine the probability of the Mössbauer effect.<sup>[3]</sup> For example, on the Debye approximation one cannot explain the low value of  $f'$  for the SnNb<sub>3</sub> crystal and the extremely high value for the SnO<sub>2</sub> crystal.

Such an approximation might be reasonable for the monatomic  $\beta$ -Sn crystal; but the known values of the Debye temperature of tin, as found from specific heat measurements, give considerably

Quantity*	SnO <sub>2</sub>	SnO	$\beta$ -Sn	SnNb <sub>3</sub>
$f'$ { 78°K	~1	0.47±0.07	0.4±0.1	0.3 ±0.1
293°K	0.7±0.1	0.15±0.05	~0.1	0.06±0.02
$\delta, 10^{-8}$ eV { 78°K	≤2	22±2	20.0±1.5	15±2
293°K	0.0	21.5 ±1.5	19.5±1.0	11±1
$\Delta, 10^{-8}$ eV { 78°K	0	12.5 ±1.5	11.0±1.5	0
194°K	—	—	8.0±1.6	—
293°K	0	11.0 ±1.5	4.6±1.5	0

\* $f'$  is the probability of recoilless absorption of  $\gamma$  quanta,  $\delta$  is the shift of the  $\gamma$  transition energy relative to the transition energy in the SnO<sub>2</sub> crystal,  $\Delta$  is the quadrupole splitting. For the SnO and  $\beta$ -Sn crystals,  $\delta$  was determined from the position of the "center of gravity" of the split line.

greater values of  $f'$  than the experimental value. Mitrofanov and Shpinel'<sup>[17]</sup> measured the temperature dependence of the resonance absorption effect for the  $\beta$ -Sn crystal; the dependence is not in agreement with that computed theoretically on the Debye approximation. Agreement could be obtained if one assumed that the Debye temperature of the  $\beta$ -Sn crystal changes from 180° at liquid nitrogen temperature to 165° at temperatures above 200° K. But these values of the Debye temperature lead to values of  $f'$  which are far greater than the value of  $f'$  found in the present work. Furthermore, the relative change in the magnitude of the effect with temperature which they found is in agreement with the change in the value of  $f'$  found in the present work.

The lattices of the  $\beta$ -Sn and SnO crystals are tetragonal, and in both crystals the tin atoms do not have spherically symmetric surroundings. In such lattices we may expect considerable electric field gradients at the tin nuclei, and this is confirmed by the observed quadrupole splitting in these crystals. The SnNb<sub>3</sub> crystal has a lattice like that of  $\beta$ -tungsten, which is very close to cubic; in such a lattice one should not find any sizable quadrupole splitting. The SnO<sub>2</sub> lattice is not cubic, but in this lattice the surroundings of the tin atom should be much more symmetric than they are in  $\beta$ -Sn and SnO, which is probably the reason for the absence of a quadrupole splitting in the SnO<sub>2</sub> crystal. Unfortunately it is not possible at present to make a sufficiently exact calculation of the electric field gradients in the  $\beta$ -Sn and SnO crystals and thus to determine the quadrupole moment of the Sn<sup>119</sup> nucleus in the excited state.

The observed changes in energy of the  $\gamma$  transition for the different crystals (the values of  $\delta$  in the table) can be explained very naturally as the result of the interaction of the nucleus with the outer electrons (mainly with s-electrons), which is different for the ground and excited states of the nucleus and changes according to the chemical surroundings of the tin atom in the crystal (the isomer shift).<sup>[9,18,19]</sup> The value of the shift should be proportional to the product  $\Delta\langle R^2 \rangle \cdot \Delta\rho_e(0)$ , where  $\Delta\langle R^2 \rangle$  is the change in the square of the effective radius of the nucleus when we go from the ground state to the excited state and  $\Delta\rho_e(0)$  is the change in the electron density in the neighborhood of the nucleus when we change the chemical environment of the tin atom in the crystal. The theoretical calculation of the interaction of the nucleus and the electrons

requires a knowledge of the electron wave functions for atoms in crystals; such calculations would enable us to get additional information concerning the charge distribution in the nucleus and its change when the nucleus is brought to the excited state.

The influence of the temperature on the hyperfine structure of the  $\gamma$  radiation and on the energy of the  $\gamma$  transition is an extremely interesting effect. When the crystal temperature changes there is a change in the amplitude of thermal vibration and a change in the interatomic spacings; as a consequence we may expect a change in the electric field gradient at the nucleus and a change in the interaction of the nucleus with the outer electrons in the crystal. Other factors, such as pressure, can also have an analogous effect. Such effects can be calculated in principle (cf., for example,<sup>[20]</sup>). A comparison of the theoretical computations with experimental data would also be very interesting for this case.

The authors thank N. E. Alekseevskii for useful discussions.

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Translated by M. Hamermesh  
233