DEPENDENCE OF THE γ-QUANTUM RESONANCE ABSORPTION SPECTRUM ON CRYSTAL TEMPERATURE

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The spectra of resonance absorption of 23.8-keV γ rays in SnO_2 are investigated in the temperature range from 78 to 645°K. Extrapolation of the observed line widths to zero absorber thicknesses leads to values which are greater than the natural width and which grow with increasing temperature. The experimental data show that the broadening is not the result of a doublet structure and that the true broadened line has an approximate Lorentz shape. The probabilities for absorption without recoil (f') are measured in the investigated temperature range. The values of f' vary weakly with the temperature. A linear dependence of the temperature line shift due to a second-order Doppler effect was observed at $T \gtrsim 300$°K. Deviations from linearity occur at $T \lesssim 300$°K.

INTRODUCTION

As is well known, the probability of recoilless absorption (emission) of gamma quanta in crystals (f'), calculated in the Debye approximation, decreases sharply with increasing temperature. It has been already noted earlier that in many cases of complicated lattices the value of f' turns out to depend little on the temperature. In resonance absorption spectra single lines broader than the natural width are frequently observed, the width being found to exhibit a temperature dependence in the case of white tin[1,2]. A change in temperature also influences the position of the resonance line and leads to so-called temperature shifts, due to the second-order Doppler effect. It is hoped that a detailed investigation of the foregoing temperature effect can yield valuable information on the dynamics of the crystal lattice.

In the present investigation we studied the resonance absorption spectra of 23.8-keV gamma rays from Sn^{119*} with an absorber in the form of polycrystalline tin oxide powder (SnO_2) at different temperatures.

MEASUREMENTS AND RESULTS

A previously described[2] setup was used to plot the absorption spectra for resonant filters of different thickness. In the experiments, the source, in the form of tin oxide 8 mg/cm² thick, was at the temperature of liquid nitrogen, while the absorber temperature ranged between 78 and 645°K. The observed spectrum had the form of a single unsplit line at all temperatures. The number of pulses counted at each observation point reached $\sim 10^5$.

Figure 1 shows the observed values of the line half-width for absorbers of different thickness at the four temperatures indicated on the plot. To determine the half-width Γ_{obs} we drew the pedestal of the line in such a way that the point of the spectrum located at $\frac{3}{2}\Gamma_{obs}$ away from the center of the line was at 10 per cent of the line height. It is seen from Fig. 1 that, within the limits of the experimental errors, the experimental points pertaining to a specified temperature lie on a straight line. The half-widths of the lines extrapolated to zero absorber thickness Γ_{extr} are listed in column 2 of the table and exceed the natural line width, the broadening increasing with increasing temperature. (Within the limits of the measurement errors, the values of Γ_{extr} are almost the same for

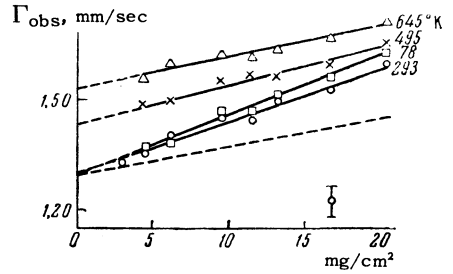

FIG. 1. Half-width of the observed spectrum vs. absorber thickness for different temperatures. Dashed line – calculated for room temperature (under the assumption of Art. 1).

T°, K	Γ_{extr}, mm/sec	Δ_A (in units of $\Gamma/2$)*	$f'_{(I)}$	$f''_{(II)}$	$\beta_A = \Gamma_A/\Gamma$
78	1.29	3.2	0.62	0.70	2.1
293	1.30	3.3	0.50	0.56	2.1
495	1.43	4.1	0.42	0.50	2.5
645	1.53	4.5	0.35	0.43	2.8

Γ – natural width of the 23.8-keV level of Sn^{119}.

78 and 293°K.) Earlier measurements of the line width made out in our laboratory[3] for the absorber, yielded for the extrapolated value of Γ_{extr} at room temperature a value close to the natural line width. This deviation from our present data is essentially due to the inaccuracy of the results obtained in [3] for thin absorbers with weak sources.

Figure 2 shows the values of the maximum absorption $\epsilon_0 = [N(\infty) - N(0)]/N(\infty)$, measured with absorbers of different thickness at room temperature. The value of f′ obtained from measurements of this type is very sensitive to the structure of the emission and absorption lines. The observed line shape differed little from Lorentzian, but the possible presence of a doublet structure in the emission and absorption lines cannot be excluded.

1. We consider first the case when the broadening is due to such a doublet structure of the emission and absorption lines, with distance Δ_S or Δ_A between components. The values of Δ_S and Δ_A were calculated by integration using the values of Γ_{extr} taken from the table. The values of Δ_A for different temperatures are listed in column 3. We have also calculated curves showing the dependence of ϵ_0 on the effective absorber thickness $C_A = n\sigma_0 f'$ for different values of Δ_A. In Fig. 2 the calculated curve I for $\Delta_A = 3.2$ comes closest to the experimental points for f′ = 0.50 and $\alpha f = 0.42$ (αf is the fraction of the total counting rate due to recoilless gamma quanta.) The values of f′ determined in this manner for various temperatures are listed in column 4 of the table. The effect of splitting on ϵ_0 can be seen by comparison with curve II of Fig. 2, calculated for the case of single unbroadened Lorentzian emission and absorption lines for the same values of f′ and αf. Using now the obtained value of f′, we can calculate the dependence of the observed line width Γ_{obs} on the absorber thickness. This dependence is shown for room temperature in Fig. 1 by a dashed curve. We see that this curve, calculated under the assumption of doublet line splitting, is not in position to explain the rapid increase in the width of the observed spectrum with absorber thickness.

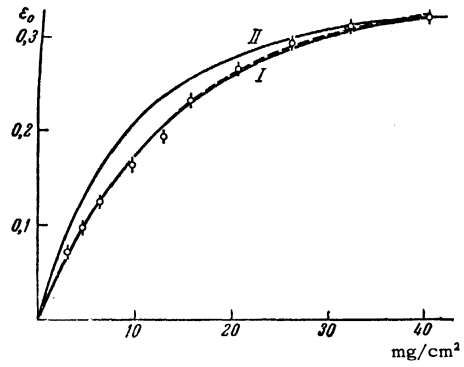

FIG. 2. Plot of maximum absorption ε_0 against absorber thickness at room temperature. Calculated curves: I – lines with doublet structure for $\Delta_A = \Delta_S = 3.2$, f′ = 0.50, $\alpha f = 0.42$; II – single unbroadened lines for the same f′ and αf; dashed line – for single broadened lines, in accordance with formula (2) with f′ = 0.56 and $\alpha f = 0.47$.

2. Let us consider the case when the emission and absorption lines are broadened, but their shape remains Lorentzian with widths Γ_S and Γ_A, respectively ($\Gamma_S, \Gamma_A > \Gamma$). For this case calculation yields the following simple expressions for the parameters of the observed spectrum.

The area of the absorption curve is

$$S = \int_{-\infty}^{\infty} \varepsilon(y)\,dy = \pi\alpha f C_A e^{-C_A/2\beta_A}\,[I_0(C_A/2\beta_A) + I_1(C_A/2\beta_A)]. \quad (1)$$

The maximum absorption is

$$\varepsilon_0 = \frac{\alpha f C_A e^{-C_A/2\beta_A}\,[I_0(C_A/2\beta_A) + I_1(C_A/2\beta_A)]}{\beta_A \varkappa(C_A/\beta_A) + \beta_S \varkappa(C_S/\beta_S)}. \quad (2)$$

The width of the observed spectrum is

$$\Gamma_{obs} = [\beta_A \varkappa(C_A/\beta_A) + \beta_S \varkappa(C_S/\beta_S)]\,\Gamma, \quad (3)$$

where β_A and β_S are equal to Γ_A/Γ and Γ_S/Γ, respectively,

$$\varkappa(\lambda) = \frac{\lambda e^{-\lambda/2}\,[I_0(\lambda/2) + I_1(\lambda/2)]}{1 - e^{-\lambda/2} I_0(\lambda/2)} - 1,$$

with I_0 and I_1 Bessel functions of imaginary argument. The dashed curve of Fig. 2 is plotted in accordance with expression (2) for $\alpha f = 0.47$ and f′ = 0.56.

The processing of the experimental data in accordance with formulas (2) and (3) leads to the results shown in columns 5 and 6 of the table. Under the same assumption concerning the shape of the emission and absorption spectra, we obtain good agreement with the experimental data for the dependence of Γ_{obs} on the absorber thickness.

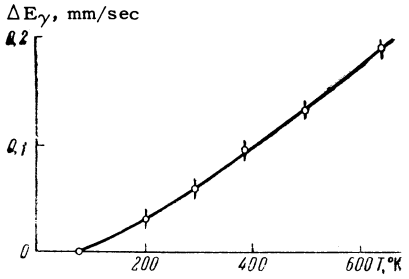

FIG. 3. Temperature shift of resonance absorption lines in SnO_2.

The results of the measurements of the temperature shift are presented in Fig. 3.

Control measurements with variation of the temperature of the source (absorber 7.8 mg/cm^2 at 293°K) confirm the previously given results.

DISCUSSION OF RESULTS

It is known that the following formula holds for the width of the second-order Doppler shift

$$\Delta E_\gamma = -E_\gamma \overline{v^2}/2c^2, \qquad (4)$$

where $\overline{v^2}$ pertains to the emitting (absorbing) nucleus. In averaging a quantity such as $\overline{v^2}$ over the spectrum of the thermal vibrations we can use for high temperatures the equipartition theorem. This leads to the following relation for the shift ΔE_γ, expressed in terms of the corresponding Doppler velocity:

$$\Delta E_\gamma\,(\text{mm/sec}) = -\frac{3}{2}\frac{kT}{M_{Sn}c} = -3.50\cdot 10^{-4}T, \qquad (5)$$

from which follows a linear dependence of the shift on the temperature. The experimental data (see Fig. 3) actually confirm the linear dependence of the shift for $T \gtrsim 300$°K and give a correct value of the slope of the straight-line portion of the curve $(3.50 \pm 0.2) \times 10^{-4}$ mm/sec-deg K. For $T \lesssim 300$°K, as can be noted from Fig. 3, deviation from linearity is observed, this being a manifestation of the well known effect of the "quenching" of the degrees of freedom at low temperatures. The experimental curve on Fig. 3 is in good agreement with the theoretically calculated curve for the Debye spectrum with end-point frequency $\Theta = 300$°K (on the temperature scale). An account of the experimental errors yields for Θ an interval of values from 200 to 350°K.

The data obtained on the basis of both assumptions concerning the line structure cannot be fitted into the framework of the Debye model. In fact, calculation in accordance with the data of column 4 of the table leads to a value $\Theta = 216$°K at $T = 78$°K and $\Theta = 367$°K at $T = 645$°K. Such a situation shows that the real phonon spectrum of the crystal SnO_2 is not of the Debye type. It must be noted that exact measurements of the temperature dependence of f' and of the temperature shift for one and the same crystal yield information concerning the real phonon spectrum.

Our results, which pertain to the observed absorption-line broadening, do not agree, as noted above, with the assumed doublet structure of the lines, which could arise, for example, as the result of quadrupole interaction. It is likewise impossible to reconcile the sign of the effect, namely the increase in line width with temperature, with such an assumption. In fact, the electric field acting on the nucleus in the lattice can be represented in the form of the sum of a static field, determined by the mean positions of the atoms, and a dynamic field, connected with the thermal motion. The change in the average positions of the atoms and of the amplitude of the thermal vibrations of the lattice should lead, as in the case of radiospectroscopic experiments on quadrupole resonance [5,6], to a reduction in the magnitude of the splitting with increasing temperature. For some tin compounds we have observed a decrease in width with increasing temperature.

One of the possible explanations of the observed line broadening is connected with the influence of the perturbation produced by the dynamic field of the lattice, which leads to natural broadening of the true absorption spectrum and consequently to a temperature dependence of the width of the observed spectrum. A consistent examination of the influence of the dynamic field with account of the finite radiation width of the nuclear levels was made by Bykov [7].

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[1] Bryukhanov, Delyagin, Opalenko, and Shpinel', JETP **43**, 432 (1962), Soviet Phys. JETP **16**, 310 (1963).

[2] Alekseevskiĭ, Pham Zuy Hien, Shapiro, and Shpinel', JETP **43**, 790 (1962), Soviet Phys. JETP **16**, 559 (1962).

[3] Delyagin, Shpinel', and Bryukhanov, JETP **41**, 1347 (1961), Soviet Phys. JETP **14**, 959 (1962).

[4] G. A. Bykov and Pham Zuy Hien, JETP 43, 909 (1962), Soviet Phys. JETP **16**, 646 (1963).

[5] Kushida, Benedek, and Bloembergen, Phys. Rev. **104**, 1364 (1958).

[6] R. R. Hewitt, Phys. Rev. **212**, 45 (1961).

[7] G. A. Bykov, JETP **44**, 249 (1963), Soviet Phys. JETP **17**, 171 (1963).

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