RESONANCE ABSORPTION OF GAMMA QUANTA IN MAGNESIUM STANNIDE. 23.8 keV ABSORPTION LINE WITH THE NATURAL LINE WIDTH

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The resonance absorption of 23.8 keV γ quanta by Sn^{119} nuclei was studied over the temperature range from 77 to 290°K in the compound Mg_2Sn. The probability of recoilless absorption of γ quanta was found to be 0.77 at 77°K and 0.28 at room temperature. The width of the absorption line in Mg_2Sn was 0.32 ± 0.02 mm/sec, which is in excellent agreement with the value from the lifetime of the 23.8 keV excited state. The results are interpreted on the basis of data concerning the structure and the nature of the chemical bonds in Mg_2Sn.

1. INTRODUCTION

WHEN there are two or more different atoms in the unit cell of a crystal the probability for recoilless resonance absorption of γ quanta and its temperature dependence show various features related to the appearance of optical branches in the vibration spectrum of the crystal.[1] In particular, in such cases there may be a high probability for recoilless absorption at high temperatures and a weak dependence of the probability on temperature. Of particular interest is the case where a heavy atom responsible for the Mössbauer effect is surrounded by lighter atoms. As was shown by Kagan,[1,2] the amplitude of oscillation of a heavy atom in an optical branch depends critically on the character of the interaction of the atoms in the lattice: optical branches play a greater role in the vibrations of the heavy atom when the interaction between the heavy atoms is stronger. But if the interaction between the heavy atoms is weak, then the heavy atom practically does not vibrate with the frequencies of the optical branches. Thus a study of the probability of recoilless resonance absorption and its temperature dependence allows one to get information about the character of the vibrations of the heavy atoms and the interaction between such atoms.

The features of resonance absorption which we have described were observed experimentally using the Mössbauer effect to study the oxides of tin,[3,4] dysprosium,[5] and europium.[6] A natural continuation of such studies is the measurement of the probability for the Mössbauer effect and its temperature dependence for a heavy atom surrounded by light atoms other than oxygen.

The object of investigation in the present study was Mg_2Sn. Its structure is antiisomorphic to the structure of fluorite CaF_2, since the more electronegative component Sn occupies the positions of the electropositive element Ca. In Mg_2Sn saturated chemical bonds are attained by exciting s electrons to p states and forming sp^3 hybrid bonds. The coordination polyhedron of the Mg atoms is a tetrahedron, while that of the tin atoms is a cube formed by eight magnesium atoms (cf. Fig. 1). Such a structure has high symmetry, so that the electric field gradient at the tin nuclei should be zero. The study of resonance absorption of γ quanta in Mg_2Sn is therefore also of interest from the point of view of finding a source of 23.8 keV γ rays with the natural width.

In our earlier investigations we usually used a source of tin dioxide, which is very convenient because of its high probability for recoilless emission at room temperature. The width of the emission line of such a source is, however, about twice the natural width of the 23.8 keV line, which spoils the resolving power of the method and complicates the treatment of the experimental results. The emission line is narrower for metallic tin, which is rarely used as a source, but in this case also the line width is much greater than the natural width; in addition, for metallic tin the probability of recoilless emission at 77°K is smaller by a

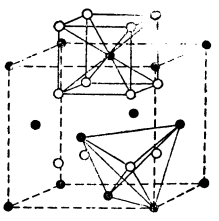

FIG. 1. Structure of Mg_2Sn (solid circles—Sn, open circles—Mg).

factor of two than for tin dioxide. It is therefore important to search for a source of 23.8 keV radiation which combines a high probability for recoilless emission with a line width which is close to the natural width.

2. DESCRIPTION OF THE EXPERIMENT

The preparation of Mg_2Sn involves no special difficulties. This compound is the only one in the binary system Mg–Sn and has a relatively low melting point, 778°C. But Mg_2Sn reacts with quartz, so the melting of the appropriate amounts of tin and magnesium was done in an iron crucible. Examination of powder patterns taken with iron radiation showed that only a single phase of the compound Mg_2Sn, with lattice parameter a = 6.764 Å, is present. Absorbers were prepared by depositing Mg_2Sn powder on aluminum foil. Measurements of the absorption spectra were made with the apparatus described earlier.[7]

Preliminary measurements were made with a tin dioxide source. These measurements showed that the width of the absorption line in Mg_2Sn is close to the natural width, and that the probability for recoilless absorption at 77°K is very high. The isomer shift of the absorption line was +(1.82 ± 0.02) mm/sec at 77°K. We then prepared a source of Mg_2Sn. Metallic tin (88.6% Sn^{118} and 1.5% Sn^{119}), irradiated with thermal neutrons in a reactor, was melted in vacuum with an amount of magnesium corresponding to the composition of Mg_2Sn. A source of thickness 12.5 mg/cm² was prepared by depositing a powder of the active Mg_2Sn on an aluminum foil.

All the later measurements were made with the Mg_2Sn source kept at 77°K. Absorption spectra were measured at room temperature and at 77°K for absorbers of 16 different thicknesses in the range from 5 to 50 mg/cm². For each absorber we determined the magnitude of the resonance absorption effect and the line width at half maximum, Γ_{exp}. The shape of the absorption line was close to Lorentzian. Measurements of the temperature dependence were made over the range from 77 to 290°K with an 8.5 mg/cm² absorber. Figure 2 shows a typical absorption spectrum obtained with a 15.3 mg/cm² absorber at 77°K.

3. EXPERIMENTAL RESULTS. CALCULATION OF PROBABILITY FOR RECOILLESS RESONANCE ABSORPTION

The probability f′ for recoilless resonance absorption was determined by two independent methods: from the dependence on absorber thickness

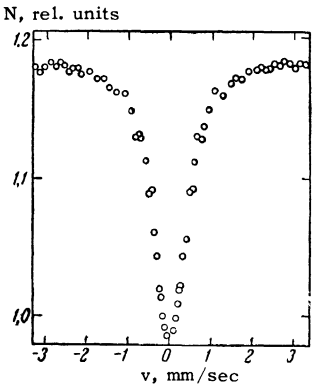

FIG. 2. Absorption spectrum for Mg_2Sn absorber, 15.3 mg/cm² thick, at 77°K. The abscissa gives the absorber velocity, the ordinate the quantum intensity in relative units.

of the line width Γ_{exp} and of the magnitude of the effect ϵ. For not too thick absorbers ($C_a \leq 5$), the dependence of Γ_{exp} on C_a can be described in the form

$$\Gamma_{exp} = \Gamma_s + \Gamma_a + 0.27\,\Gamma_a C_a, \qquad (1)$$

where Γ_s and Γ_a are the emission and absorption line widths respectively, $C_a = \sigma_0 n f' \Gamma/\Gamma_a$ (where Γ is the natural width, n is the number of nuclei per cm² of absorber of the isotope responsible for the Mössbauer effect, and σ_0 is the maximum cross section for resonance absorption). In formula (1) we can introduce explicitly the usual absorber thickness a in mg/cm² using the proportionality $C_a = Ka$. Then a comparison of (1) with the experimental data allows a determination of $\Gamma_s + \Gamma_a$ and the proportionality constant K.

The corresponding experimental results (for an absorber at room temperature) are shown in Fig. 3.

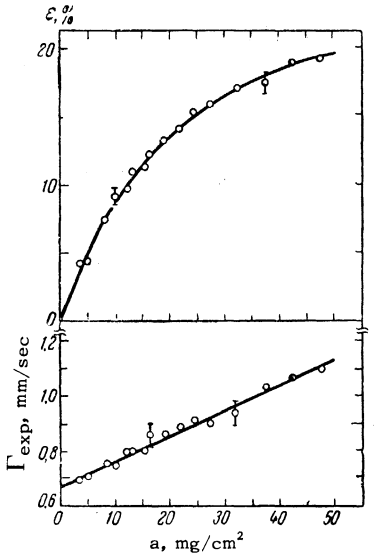

FIG. 3. Dependence of resonance absorption effect ϵ and observed line width Γ_{exp} on absorber thickness a for an Mg_2Sn absorber at room temperature. The curves were calculated theoretically using the parameter values given in the text.

Within the limits of accuracy of the measurements, the values of $\Gamma_s + \Gamma_a$ are the same at 77 and 290°K, and equal to 0.68 ± 0.01 mm/sec. The value of K was 0.11 ± 0.02 at room temperature and 0.30 ± 0.05 at 77°K. Using these data and knowing the source thickness in the Sn^{119} isotope, we found a value of 0.36 ± 0.01 mm/sec for Γ_s, which gives a value of 0.32 ± 0.02 mm/sec for Γ_a, in excellent agreement with the value from the lifetime of the 23.8 keV excited state.[8] Thus the 23.8 keV absorption line in Mg_2Sn has the natural width.

Using the results of various papers (cf. [4,9]), we can write the resonance absorption effect in the form

$$\varepsilon = \varkappa f \frac{\gamma K(C_a)}{\gamma U(C_a)+1}, \qquad (2)$$

where $\gamma = \Gamma_a/\Gamma_s$, f is the probability for recoilless emission from the source, κ is the fraction of γ radiation with energy 23.8 keV in the total flux of quanta recorded by the detector; the functions $K(C_a)$ and $U(C_a)$ are defined in the papers cited above.[4,9] Since the parameter γ is known, comparison of computed curves of the type of (2) with the experimental data again permits a determination of the constant K. This method gave values of K of 0.093 ± 0.007 at room temperature and 0.27 ± 0.02 at 77°K, which agree with the values determined above from the dependence of Γ_{exp} on absorber thickness. In the computation we used average values of 0.10 ± 0.01 at room temperature and 0.28 ± 0.02 at 77°K. Using the values found for K, we find for the probability of recoilless resonance absorption of 23.8 keV γ quanta in Mg_2Sn the following values: f'(77°) = 0.77 ± 0.08 and f'(290°) = 0.28 ± 0.03. (The estimates of error of f' include only the error in our measurements. Errors in the measurements of the lifetime and the conversion coefficient[8] could increase the overall uncertainty in the determination of the absolute values of f').

The dependence of the resonance absorption effect on temperature was measured over the range from 77 to 290°K. Since there was no noticeable change of Γ_a with temperature, the computation of the values of C_a and f' from the magnitude of the effect could be done using the dependence of ϵ on absorber thickness at room temperature (or at liquid nitrogen temperature). The temperature dependence found for f' is shown in Fig. 4.

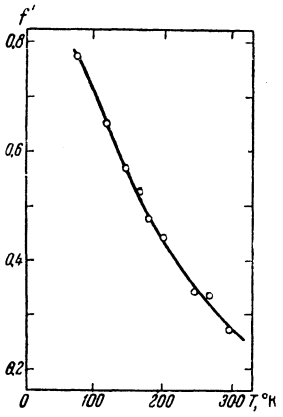

FIG. 4. Temperature dependence of probability of recoilless absorption of 23.8 keV γ quanta by Sn^{119} nuclei in Mg_2Sn.

4. DISCUSSION OF RESULTS

As was already mentioned, in Mg_2Sn the predominant interaction is between tin and magnesium atoms. In that case the contribution of optical vibrations for the heavy tin nucleus responsible for the Mössbauer effect is considerably reduced. Only at quite low temperatures, when the total number of phonons is small, will the role of the optical vibrations become important. With increasing temperature the probability f' drops rapidly because of the excitation of acoustic phonons. This corresponds to a high value of f' at 77°K and a rapid drop with increasing temperature. The characteristic temperature computed from the experimental value of f' at room temperature was 200°. This value is in good agreement with the characteristic temperature of 201° found for Mg_2Sn by the x-ray method,[10] and with the value of 206°, calculated in the same paper[10] from the formula

$$\theta = 19.37/\sqrt{\bar{A}V^{1/3}\alpha},$$

where $\bar{A}$ is the mean square value of the atomic weight, α is the linear expansion coefficient and V is the atomic volume.

It is of interest to use the Mössbauer effect to study compounds with a structure isomorphic to fluorite ($PtSn_2$, $IrSn_2$). These compounds have metallic binding, so that we may expect tighter binding between atoms of the same kind and, consequently, an increased effect of the optical vibrations for the tin atoms.

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