ENERGY SPECTRUM OF RESONANCE ABSORPTION IN ZINC OXIDE OF THE 92-keV *γ RADIATION FROM* Zn^{67}

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Submitted to JETP editor December 9, 1961

J. Exptl. Theoret. Phys. (U.S.S.R.) 42, 1029-1035 (April, 1962)

Experiments are described on the observation of the Mössbauer effect for the 92-keV γ radiation from Zn^{67}, using a source and filter of zinc oxide. Both Doppler shift and frequency-modulation methods were used. The magnitude of the resonance effect reaches a value of 2×10^{-3}, while the line width is somewhat greater than the natural width. The spectrum shows a structure which is difficult to interpret with data of the present accuracy.

1. INTRODUCTION

THE first excited state of Zn^{67} has an energy $E_0 = 92$ keV and a half-life of $T_{1/2} = 9.6 \times 10^{-6}$ sec (natural width $\Gamma = 4.9 \times 10^{-11}$ eV); it can be obtained from K capture in Ga^{67} ($T_{1/2} = 78$ h) or β decay of Cu^{67} ($T_{1/2} = 67$ h).[1]

The study of the Mössbauer effect[2] for the 92-keV radiation arising from the transition from this state to the ground state is of considerable interest since the Mössbauer effect in zinc can, in principle, be used for many interesting experiments, as for example:

1. Detection of very small changes in frequency of the γ radiation (since the relative width of this γ ray line in Zn^{67} is $\Gamma/E \approx 5 \times 10^{-16}$, which is three orders of magnitude smaller than the relative width of the Fe^{57} line with which the gravitational shift was measured.[3]

2. Measurement of the magnetic and quadrupole moments of the Zn^{67} nucleus in its first excited state.

3. Measurements of certain characteristics of solids containing Zn atoms in their lattice (internal fields, isomeric shifts, etc).

However the study and use of the Mössbauer effect in Zn^{67} is complicated by several facts. The relatively large recoil energy of the nucleus after the γ transition results in a small probability for "recoilless" emission and absorption.

Because of the small width of the 92-keV line of Zn^{67}, there are difficulties with all sorts of splittings or broadenings of the emission and absorption line, which may make the Mössbauer effect in Zn^{67} practically unobservable. Some of the possible causes of such splittings and broadening are: 1) presence of magnetic and inhomogeneous electric fields at the positions of Zn atoms; 2) perturbations from decays preceding the formation of the excited state; 3) incorrect location of some of the Zn^{67} atoms in the lattice of the solid; 4) relative vibrations of source and filter.

Several papers have appeared in the literature on the observation of the Mössbauer effect for Zn^{67}. All of these were done by the transmission method using the parent nucleus Ga^{67}.

The Mössbauer effect in Zn^{67} was not seen in the experiments of Pound and Rebka,[4] which they did for various alloys and compounds of Zn, to an accuracy of 0.1%.

Aksenov et al.[5] using a metallic source and filter (the filter was enriched to 33% in the Zn^{67} isotope and had a thickness ~ 1.5 g/cm^2) at a temperature of 4.2°K obtained an increase of 0.03% in the transmission of the absorber when the resonance condition was destroyed by a magnetic field.

A considerably larger effect was obtained in the work of Craig et al,[6] where the source and absorber were samples of ZnO cooled to 2°K. They measured the transmission of the absorber as a function of the applied magnetic field (the filter was enriched to 92.4% Zn^{67} and had a thickness 1.2 g/cm^2). The maximum change in transmission observed was 2.5×10^{-3}. A structure was observed in the field dependence of the transmission, the maximum resonance absorption corresponding to a field of ~ 10 Oe. The interpretation of the results[6] is made difficult by the low accuracy of the measurement and the lack of knowledge of the magnetic moment of the excited state in Zn^{67}.

We present here the results of an investigation of the Mössbauer effect in Zn^{67}, also done by the transmission method using the parent nucleus Ga^{67}, but using the Doppler shift to obtain the energy spectrum of the resonance absorption.

2. EXPERIMENTAL ARRANGEMENT

The energy spectra of the resonance absorption of γ quanta were obtained by measuring the dependence of the transmission of the filter for the 92-keV line of Zn^{67} on the relative velocity of source and absorber.

The principal measurements were done with source and absorber cooled to ~ 4.2°K. The source was a thin foil (20 × 10 × 1 mm) of baked natural ZnO, irradiated with a beam of 13.4-MeV deuterons. As a result of the irradiation and the resulting $Zn^{66}(d,n)Ga^{67}$ reaction, an activity of several tens of millicuries of Ga^{67} is produced on the foil surface. To eliminate radiation damage to the crystal lattice, the source was annealed in air for 1—2 hours at 1000—1100°C.

The absorber was made in the form of a thin-walled brass container, filled with ZnO powder bonded with polyvinyl alcohol. The Zn was enriched to 33% in the Zn^{67} isotope. The absorber thickness was ~ 1.6 g/cm^2.

A schematic diagram of the experimental arrangement is shown in Fig. 1.

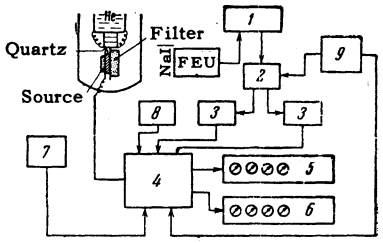

FIG. 1. Block diagram of the experimental equipment: 1) single-channel analyzer, 2) fast commutation circuit, 3) counting circuit, 4) slow commutation circuit 5) scaler for positive speeds, 6) scaler for negative speeds, 7) sound generator, 8) timer, 9) generator of sawtooth pulses.

To the helium vessel of the cryostat, which sat on a felt support (to isolate the source and absorber from external sources of vibration), there was attached a copper platform, connected to the helium dewar by flexible copper heat pipes. On one side of the platform there was soldered a container with the filer, while a piezo-quartz plate (X-cut, thickness 2.5 mm) was fixed to the other side with Ramsey cement. The source, with a thin copper foil between, was in turn attached to the piezo-quartz, with the irradiated face toward the absorber.

Gamma radiation passing through the absorber was recorded by a scintillation counter using a thin (2.5 mm) crystal of NaI. The 92-keV line was selected from the pulse spectrum by a single-channel AADO-1 analyzer.

To obtain an energy spectrum of the resonance absorption, a triangular (sawtooth) voltage pulse was applied to the quartz, with a frequency of 1.5 kc and an amplitude which ran periodically through a number of fixed values from 0 to U_{max} (where U_{max} reached 1500 V in some measurements). To each value of the amplitude of the triangular voltage pulse there correspond two relative velocities $\pm v$, equal in magnitude but opposite in sign, which cause an energy shift $E_0 v/c$ between the emission spectrum of the source and the absorption spectrum of the absorber. The fast commutation circuit, controlled from the generator of the triangular voltage pulses, enabled us to record in two different counting channels the counts corresponding to the energy displacements of opposite sign. This same circuit also shut off the counting channels for a few tens of microseconds in the neighborhood of the reversals of the triangular voltage. The abrupt changes in amplitude of the triangular voltage pulses, together with the simultaneous switching between the pair of counting channels, was carried out automatically by a slow commutation circuit over equal time intervals (~ 10 sec), set by an interval counter which was quartz stabilized. The contribution to the energy spectrum from the change in intensity of the source due to decay of the Ga^{67} was eliminated with sufficient accuracy for our case by suitable adjustment of the channels in successive cycles of the triangular voltage pulses.

In the measurements with frequency modulation of the γ radiation,[7] a sinusoidal voltage of low frequency and amplitude was applied to the quartz oscillator through a slow commutation circuit.

In order to determine the transmission of the absorber when resonance absorption is definitely absent, in each cycle there was a reference channel during which a sinusoidal voltage of high frequency and amplitude (100 kc, 210 V) was applied to the quartz, destroying the resonance by frequency modulation.

The apparatus described enabled us after several days running to obtain a spectrum with 50 points, with a statistical accuracy at each point of 2×10^{-4}.

3. RESULTS OF MEASUREMENTS

Typical energy spectra of the resonance absorption* in ZnO for the 92 keV line, at a temperature of ~ 4.2°K for various sources, obtained with irradiation of identical samples, are shown in Fig. 2.

The calculation of the energy shifts was made using the tabulated value of the piezomodulus of quartz at 4.2°K, $d_{11} = 2.02 \times 10^{-10}$ cm/V.[8] The experimental calibration of the displacement scale will be discussed when we consider the results.

In all the energy spectra, the maximum resonance absorption occurred at zero energy shift.

The magnitude of the effect at the central peak reaches 2×10^{-3}, and varies somewhat from one source to another. After correcting for "dilution" of the γ spectra by nonresonant radiation and for resonance scattering in the detector, the total resonance effect is raised by a factor of two. The peak widths are several times the ideal width 2Γ, and

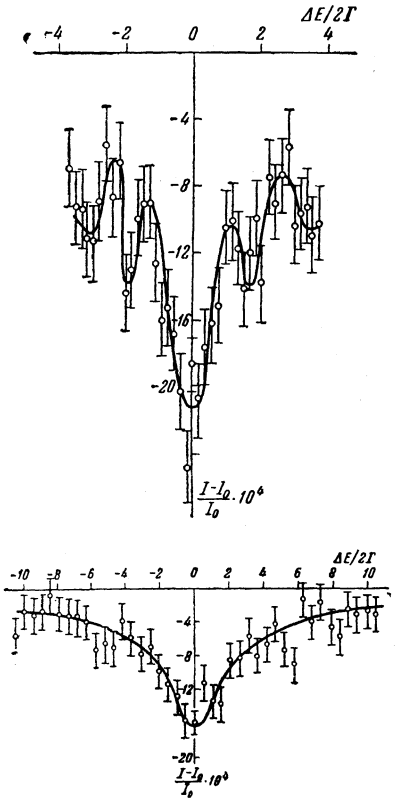

FIG. 2. Energy spectra of resonance absorption in ZnO, cooled to ~ 4.2 °K, for the 92 keV γ radiation of Zn^{67} (for various sources). The abscissa gives the energy shift (in units of 2Γ).

*By the resonance absorption energy spectrum we mean the dependence of $(I-I_0)/I_0$ on the energy shift $E_0 v/c$ between emission and absorption spectra (I and I_0 are the counting rates in the particular channel and in the reference channel, v is the speed of the source relative to the absorber, and E_0 is the energy of the γ radiation.

sources which give a smaller effect have a wider line. These variations cannot be attributed to differences in annealing conditions, since several successive annealings of one of the sources which gave a small effect made no significant change in the picture. In the spectra where the effect was larger (Fig. 2), there is an indication of a complicated structure of the resonance absorption.

The control spectrum shown in Fig. 3 was obtained with source and absorber at ~ 80°K, where the resonance absorption should be reduced considerably. Within the limits of error of the measurements, the data of Fig. 3 show no dependence of the transmission on the energy shift.

Figures 4 and 5 show the results of measurements made using frequency modulation of the 92 keV γ rays from Zn^{67}.

Figure 4 gives the dependence of the magnitude of the resonance absorption effect $(I-I_0)/I_0$ on the amplitude of the sinusoidal voltage of frequency ν = 80 kc applied to the quartz; the data are normalized to unity at U = 0. The curve of Fig. 4 was calculated using the resonance absorption spectra shown in Figs. 5 and 6, on the assumption that the 1.5 kc triangular voltage pulse is equal to the tabulated value and that for the sinusoidal 80-kc voltage it is twice as great (see the discussion of the results).

Figure 5 gives the resonance absorption spectrum of frequency-modulated 92-keV radiation; in this experiment, in addition to the scanning trian-

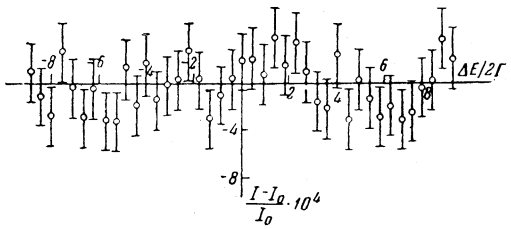

FIG. 3. Energy spectrum of resonance absorption at ~80 °K.

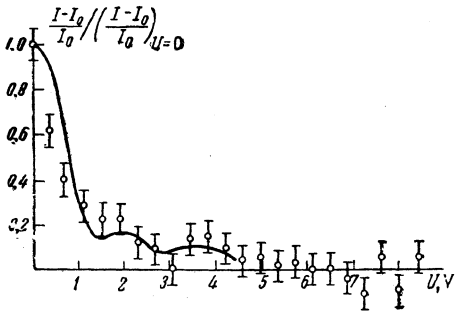

FIG. 4. Dependence of normalized ratio $(I-I_0)/I_0$ on amplitude U of the sinusoidal voltage applied to the piezooscillator.

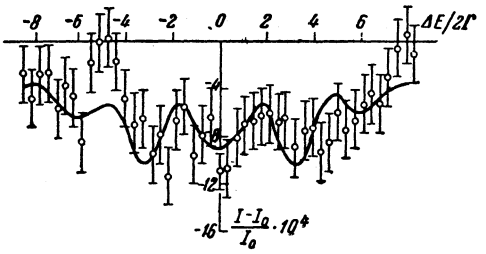

FIG. 5. Energy spectrum of resonance absorption of frequency-modulated γ radiation.

gular voltage, a sinusoidal voltage of frequency 80 kc and a constant amplitude of 1.1 V was applied to the piezoquartz; the curve is computed on the same assumptions as for Fig. 4.

For one of the sources, in addition to the resonance absorption spectrum for the enriched absorber, we also measured the resonance absorption spectrum for an absorber of normal composition with the same weight thickness (so that it was eight times thinner in Zn^{67}). The results of these measurements are shown in Fig. 6.

4. DISCUSSION OF RESULTS

The quartz oscillator which we used for producing the energy shifts requires calibration,

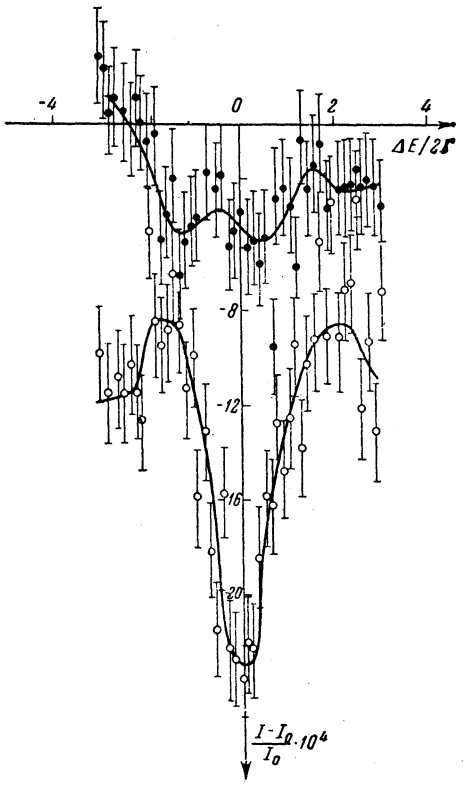

FIG. 6. Energy spectra of resonance absorption of γ rays by one of the sources for absorbers of enriched and normal composition with the same weight thickness.

since coupling of the quartz on one of its planes and possible bending oscillations may alter the effective value of the piezomodulus. To measure the actual value of the piezomodulus of the quartz under operating conditions (a shift of the order of a fraction of a millimicron in the vibrator placed in the cryostat) is extremely difficult by any known method. To calibrate the oscillator we therefore made use of the Mössbauer effect in Zn^{67}.

The lower limit of the effective value of the piezomodulus is obtained directly from the energy spectrum, if we assume that the width of the central peak of Fig. 2 is the minimum possible, i.e., is equal to twice the natural width of the level. This limit is approximately half as great as the tabulated value of the piezomodulus.

For a precise determination of the modulus, one can use the measurements of the resonance absorption with frequency modulation of the γ rays.

In frequency modulation, because of the motion of the source according to the law $x = Ud_{11} \sin \Omega t$, the spectrum of γ radiation is split into two components, whose energy displacements and relative intensities are given by the expressions

$$\Delta E = \hbar\Omega,$$

$$I_0 : I_1 : I_2 : \ldots = J_0^2(Ud_{11}k) : J_1^2(Ud_{11}k) : J_2^2(Ud_{11}k) : \ldots,$$

where U is the amplitude of the sinusoidal voltage applied to the quartz, J is the Bessel function of the first kind, and K is the wave number of the γ radiation.

If the components of the split line do not overlap and the filter is at rest, only the zeroth component suffers absorption, i.e., when the modulation is switched on the absorption decreases like $J_0^2(Ud_{11}k)$. It is easy to use this dependence to measure the magnitude of $d_{11\,\mathrm{eff}}$; however if the modulation frequency approaches the natural frequency of the vibrator, the effective value of the piezomodulus may be different from the value we are interested in, which corresponds to the frequency of the triangular voltage pulses (1.5 kc). It is therefore more convenient to calibrate the vibrator from the separation between the components of the frequency splitting in the energy spectrum of the resonance absorption of the frequency-modulated γ radiation.

We used both methods of calibration; the results are shown respectively in Figs. 4 and 5. Unfortunately the interpretation of these experiments is made difficult by the fact that, for the modulation frequency used (80 kc), the components of the frequency splitting still overlap con-

siderably because of the large width of the resonance absorption spectrum of Zn^{67}. It was difficult to increase the modulation frequency—this required a corresponding increase of the amplitude of the scanning voltage and in the number of recording channels.

In analyzing the data of Figs. 4 and 5, we used the resonance absorption spectra obtained for the same source without frequency modulation.

The best way to explain the data of Figs. 4 and 5 is to assume that for the triangular voltage of frequency 1.5 kc the piezomodulus is approximately equal to the tabulated value, while it is twice as great for the 80 kc sinusoidal voltage. The curves drawn in Figs. 4 and 5 were made on these assumptions. We see that the deviations between the experimental points and the curve in Fig. 4 are sizable. The reason for this discrepancy is not clear to us.

We note that Fig. 4 clearly shows the possibility of using the resonance absorption of the 92-keV γ radiation from Zn^{67} for measuring extremely small ($\sim 10^{-9}$—10^{-10}cm) amplitudes of mechanical vibration with frequencies of several tens of kilocycles or higher.

Craig et al[6] in their experiments on Zn^{67} observed a sizable shift between emission and absorption spectra and attributed it to a difference in zero-point vibrations of the crystal lattices of the source and absorber, resulting from the difference in their isotopic composition.

In our measurements the maximum resonance absorption occurred for zero relative velocity of source and absorber. This indicated that such a shift is either absent or small, although the average isotopic composition was almost the same in our experiments and in [6].

The structure of the resonance absorption spectra is apparently caused by the quadrupole splitting of the level of Zn^{67} in the distorted cubic lattice of ZnO.[9] To decipher the structure one would have to increase the accuracy of the measurements considerably.

The analysis of the energy spectra of resonance absorption for two absorbers with different thicknesses of Zn^{67} enabled us to evaluate the probability of "recoilless" absorption and emission of γ rays by Zn^{67}. Assuming a quadrupole splitting, we found a value of $f' = 2 \times 10^{-2}$ for the absorption probability. The emission probability f appears to be 3—5 times smaller, which may be due to a longlasting disturbance of the electronic shells of Zn^{67} formed as a result of K capture in Ga^{67}.[10] From this point of view it is of interest to measure the resonance absorption in Zn^{67} from the parent nucleus Cu^{67}, since the perturbation of the electronic shells should be considerably smaller for the β decay.

An evaluation of the effective Debye temperature Θ_D for ZnO from the value of f' gives $\Theta_D \approx 300°K$, in agreement with the results of [6].

The low value of the Mössbauer effect for Zn^{67} in ZnO complicates its use and compels us to look for ways of increasing it or making it stand out more.

In conclusion the authors thank P. A. Bazhulin for preparing the piezoquartz; A. B. Fradkov and M. P. Malkov for liquefying the helium; A. F. Ryabov, A. I. Sekirin, and A. V. Sokolov for help with the measurements, and also take this opportunity to express their deep gratitude to S. S. Vasil'ev, A. F. Tulinov, Yu. A. Vorob'ev, and the personnel of the cyclotron of the Institute for Physical Problems of Moscow State University.

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Translated by M. Hamermesh
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