MÖSSBAUER EFFECT OF THE 93-keV TRANSITION IN Zn^{67} †

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Mössbauer spectra obtained with carefully annealed sources consisting of Ga^{67} in $Zn^{66}O$ and with $Zn^{67}O$ absorbers show a well-resolved hyperfine structure that can be attributed to equal quadrupole splittings in source and absorber, with coupling constants $e^2qQ = 2.47 \pm 0.03$ MHz. The resonant absorption for the strongest line of the spectrum ranges from about 0.15 to 0.55% and its linewidth from 2.7 to 5 times the natural width $2\Gamma_0$. A pressure of about 40 kbar applied to the source gives rise to about an 8% reduction in its quadrupole coupling constant.

The 93-keV level of Zn^{67}, with $T_{1/2} = 9.6$ μsec, has a relative natural width $\Gamma_0/E_\gamma = 5.2 \times 10^{-6}$, about 600 times smaller than that of the 14.4-keV level of Fe^{57} and 20 times smaller than that of the 6.25-keV level of Ta^{181}. Comparatively little work has been done on the Mössbauer effect of this level; in the investigations reported so far a considerable effort was involved in observing the resonance at all.[1-5] Velocity spectra were first shown in the paper by Alfimenkov et al.[5] The present measurements improve and extend their work enough to permit an unambiguous interpretation of the spectra.

In the study reported here, clear spectra were observed with linewidths from 3 to 5 times the minimum possible width $2\Gamma_0 = 3.1 \times 10^{-4}$ mm/sec and an absorption dip after background correction of 0.2-0.55% for the central line of the spectrum. For the ZnO sources and absorbers used in the investigation, a well-resolved hyperfine structure was observed.

The measurements were performed with a piezoelectric velocity drive consisting of a stack of ten 6-mm-thick X-cut quartz-crystal rings of o.d. 3.8 cm and i.d. 1.9 cm. Between the crystals, thin gold electrodes are placed. The crystals are stacked with alternating polarities and the electrodes are alternately connected to ground and to a sinusoidal ac voltage. In this manner, the total piezoelectric displacement for a given voltage is 10 times that of one crystal. Source and absorber are mounted in holders that are rigidly fixed to the opposite ends of the quartz stack.

The operation of the drive was tested at room temperature by observing the intensity modulation on the steep flank of a Mössbauer absorption line (obtained with a Co^{57}-in-Cu source and a ferric sulphate absorber) for different drive voltages. The velocity calibration obtained in this way agrees within 10% with that calculated from the piezomodulus of the quartz. For the measurements at liquid-helium temperature, we therefore felt confident in basing the calibration on the known piezomodulus[6] ($d_{11} = 2.02 \times 10^{-10}$ cm/V) of X-cut quartz at 4.2°K.

The sources used for our experiments consisted of sintered disks of about 100-mg ZnO enriched to 90% in Zn^{66}, irradiated with 5-15 μA-h of 12-MeV deuterons or 30-MeV He^3 particles to yield the 70-h activity of Ga^{67}. The disks were annealed after irradiation. The absorbers were disks of 0.6-2.2 g/cm^2 ZnO enriched to 90% in Zn^{67}. All materials were obtained from Oak Ridge.

The drive assembly is suspended by nylon strings in a stainless-steel container filled with helium gas under reduced pressure and immersed in liquid helium. By using the system as a piezoelectric microphone, we could check that mechanical vibrations reaching the drive as a result of boiling helium were always far below the permissible level.

Measurements were made to determine (1) the quadrupole splitting in ZnO, (2) the influence of source and absorber preparation on the width and depth of a resonance, (3) the second-order Doppler shift, and (4) the influence of pressure on the source.

(1) Quadrupole splitting in ZnO.—In Fig. 1, spectra obtained with ZnO sources and absorbers are shown for two different velocity ranges. They exhibit a hyperfine structure that can be ascribed to equal quadrupole splittings in source and absorber. The 93-keV transition is known to occur between spin-$\frac{1}{2}$ and spin-$\frac{5}{2}$ levels and to have pure $E2$ character.[7] In this case, the source and absorber lines are split into three components of equal intensity, and we must expect a seven-line spectrum. At the highest velocity range realized so far, five of these lines are observed (Fig. 1, bottom). For the central line,

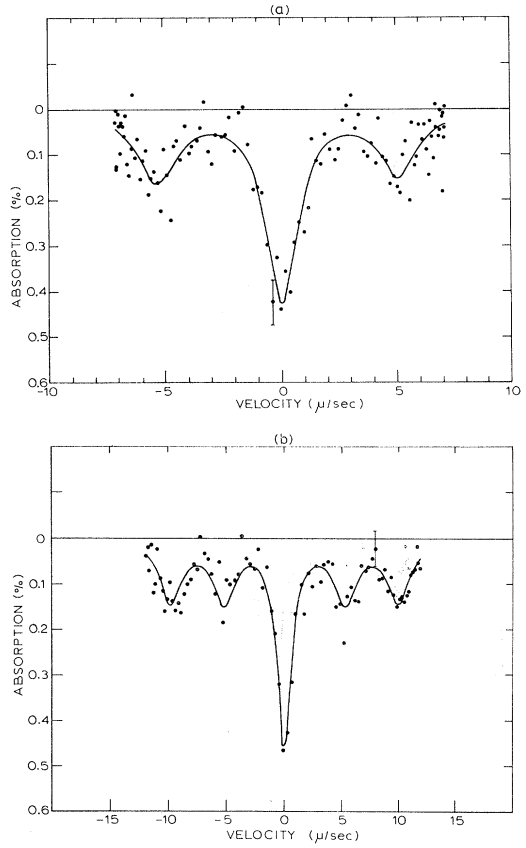

FIG. 1. $Zn^{67}O$ Mössbauer spectra for two velocity ranges: (a) from −7 to +7 μ/sec, (b) from −12 to +12 μ/sec. Source: 70-h Ga^{67} activity produced in $Zn^{66}O$ by (d,n) reaction. Absorber: 2.11-g/cm^2 $Zn^{67}O$ enriched to 89.6%.

each of the three source components overlaps with one of the absorber, while for each of the other lines only one source component overlaps with one absorber component. Therefore, the expected intensity ratio is 1:1:3:1:1, which is consistent with our experimental findings. From the positions of the lines in the Mössbauer spectrum, a quadrupole coupling constant $e^2qQ = 2.47 \pm 0.03$ MHz and an asymmetry parameter $\eta = 0.23 \pm 0.06$ are derived. Using the value[8] $Q = 0.17$ b given for the ground-state quadrupole moment of Zn^{67}, we find that the absolute value of the z component of the field gradient at the Zn^{67} nucleus is $|eq| = 6.0 \times 10^{16}$ V/cm^2.

It is interesting to compare this value with the field gradient derived for a lattice of point charges, computed from the expressions given by deWette[9] for the case of a hexagonal lattice. If one uses the most recent values[10-12] of the atomic positions ($u = 0.3826 \pm 0.0007$, $a = 3.242$ Å, $c = 5.193$ Å) and an interpolated value of the Sternheimer factor[13] ($\gamma_\infty = -12.2$) one obtains $eq_{calc} = (5.4 \pm 1.1) \times 10^{16}$ V/cm^2. This is somewhat better agreement than might have been expected in view of the fact that contributions of higher multipoles to the lattice sum are neglected. The agreement lends support to our explanation of the cause of the splitting.

The asymmetry parameter can be explained by an exceedingly small (~0.001 Å) deviation of oxygen atoms from the ideal tetrahedron.

(2) Influence of source and absorber preparation on resonance width and depth.—Though a more systematic investigation of this matter must still be made, we report some of our first observations here. Typical results for width and depth of the central line of spectra obtained with ZnO sources and absorbers prepared in various ways are shown in Table I. The strongest absorption so far has been obtained by irradiating the source with deuterons, annealing source and absorber for a few hours at 750-800°C, and then slowly cooling them (at a rate of about 30°C/h). For such materials, the linewidths obtained were 3-5 times the natural width $2\Gamma_0$. Seemingly small changes in preparation may have a large influence. For instance, an absorber that was ground to a coarse powder after sintering (Expt. 1, Table I) yielded a broad and shallow line. For a source irradiated with He^3, a broad and shallow line was found after the source had received its usual heat treatment (Expt. 5). The linewidth appears to be reduced by a second heating to a higher temperature (Expt. 6), but the depth is still only one half of that found for a deuteron-irradiated source.

The line broadening due to absorber thickness may be obtained after making an estimate of the characteristic temperature θ_m for the Zn atoms in ZnO. With $\theta_m \approx 290$°K deduced from Ref. 10, we get $f \approx 0.015$ for the recoilless fraction and a broadening $\Delta\Gamma \approx 0.2$ μ/sec—which is insufficient to account for our results. Small fluctuations in the splitting in the source or in the absorber or in both could give the observed broadening.

(3) Second-order Doppler shift.—Different isotopic compositions of source and absorber in principle give rise to a second-order Doppler shift (ΔE_D) between the resonant energies of source and absorber. Lipkin[14] had derived a general expression for this shift [his Eq. (23)] in the case of an impurity atom of mass m_0 imbedded in a monatomic lattice of slightly different mass m ($\delta m = m_0 - m \ll m$). From Lipkin's formula we find for a Debye solid that $\Delta E_D = 3.4 \times 10^{-3} (\delta m /$

Table I. Widths and intensities of central line of Mössbauer spectra obtained with various Ga^{67}-in-$Zn^{66}O$ sources and with various $Zn^{67}O$ absorbers.

Expt. No.	Source	$Zn^{67}O$ absorber	Width of central line (μ/sec)	Intensity (%) (background corrected)
1	$Zn^{66}O(d,n)$ annealed	0.66 g/cm^2 powdered	2.7 ±2	0.16 ±0.03
2	$Zn^{66}O(d,n)$ annealed	0.71 g/cm^2 sintered	0.8 ±0.15	0.19 ±0.02
3	$Zn^{66}O(d,n)$ annealed	2.17 g/cm^2 sintered	1.1 ±0.1	0.39 ±0.02
4	$Zn^{66}O(d,n)$ annealed and slowly cooled	2.11 g/cm^2 sintered and slowly cooled	1.52 ±0.12	0.55 ±0.02
5	$Zn^{66}O(He^3,pn)$ annealed and slowly cooled	2.11 g/cm^2 sintered and slowly cooled	2.1 ±0.4	0.20 ±0.02
6	Reheated to 950°C and rapidly cooled	2.11 g/cm^2 sintered and slowly cooled	0.8 ±0.3	0.25 ±0.06

$m)(k\theta_D/m_0c^2)E_\gamma$. In our measurements, the emitting nuclei are Zn^{67} in a $Zn^{66}O$ lattice; the absorbing nuclei are Zn^{67} in a highly enriched $Zn^{67}O$ lattice. This case is actually more complicated than that considered by Lipkin because the lattice is diatomic; each zinc is surrounded by an oxygen tetrahedron that has the same isotopic composition for source and absorber. Thus, if we insert $\delta m = -1$, $m = 66$, and $\theta_D = 290$°K in the expression for ΔE_D, we must regard the resulting $\Delta E_D \approx -0.006$ μ/sec as a maximum value. From our experiments 3 and 4, we derive a shift $\Delta E = 0.027 \pm 0.02$ μ/sec for the central line. Clearly, this is not sufficiently accurate to draw any conclusion about the possible presence of a second-order Doppler shift.

(4) Influence of pressure on the source.—A $Zn^{66}O$ source that had been annealed after deuteron irradiation was placed in a $\frac{1}{8}$-in.-bore stainless-steel cylinder and compressed by a screw to a pressure roughly estimated to be 40 kbar. As compared with a three-line spectrum from the same source at zero pressure, a three-line spectrum taken with this source shows (a) a shift of about -0.11 μ/sec for the central line of the spectrum, (b) a (4 ± 2)% reduction of the splitting between the two outer lines, (c) about a 25% broadening of the central line, and (d) a reduction of the ratio of center-line to outer-line intensity from 3.6 ± 0.5 at zero pressure to 2.4 ± 0.3 at high pressure. It can be shown that all these changes are compatible with about an 8% reduction of the source quadrupole splitting as a result of compression. Such a change is not unexpected if we realize that a pressure of 40 kbar gives a 3% compression of the lattice, i.e., an average reduction of about 0.04 Å in the distance between lattice atoms. Only a small compression anisotropy is needed to yield differences in the shift of the order of 0.001 Å. As seen from result (a) above, such shifts suffice to bring about the observed change of field gradient.

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THICKNESS OF THE STATIC AND THE MOVING SATURATED He II FILM*

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We have used a three-terminal capacitor incorporated in a flow vessel to search for the theoretically predicted difference in the thickness Δd between the moving and the static saturated He II film. The observations indicate that $\Delta d = 0$ to within a few Å.

Despite the considerable amount of effort that has been applied to determinations of the thickness d of both static and moving He II films,[1-3] the relation between theoretical predictions and experimental results in these matters remains unsatisfactory. An important example of this confusion concerns the difference in thickness Δd between the static and the moving film when other variables, such as the temperature and the vertical distance Z of the film above the liquid surface, are kept constant.

According to the now widely accepted ideas of Kontorovich,[4] the following expression holds in regions of the moving film where no dissipation occurs:

$$(\rho_s/2\rho)v_s^2 + gZ - \gamma d^{-n} = 0, \quad (1)$$

where ρ_s/ρ is the superfluid fraction of the total density ρ, v_s is the superfluid velocity, g is the gravitational constant, and γ is the coefficient of the van der Waals attractive potential between the helium and the substrate in which n is usually taken as 3. For the static film ($v_s = 0$) we find from Eq. (1)

$$d = (\gamma/gZ)^{1/n}, \quad (2)$$

but when $v_s \neq 0$, the Kontorovich relation indicates that the film should be thinner by an amount

$$\Delta d = (1/n)(\rho/2\rho_s)(Q^2/gZd). \quad (3)$$

Here $Q = (\rho_s/\rho)v_s d$ is the flow rate per cm of container perimeter. Experiments using optical techniques by Gribbon and Jackson[2] (flow from a beaker) and by Pickar and Atkins[3] (third sound) have indicated Δd to be smaller than expected from Eq. (3), but scatter in the data has precluded a quantitative evaluation of Δd.

The main purpose of the investigation reported here was to devise a new method for measuring Δd more precisely than in previous work. During the course of the experiments additional features of the He II film became clarified.

As a probe of the film thickness a three-terminal capacitor (nominal capacitance ~19.5 pF) was constructed as shown schematically in Fig. 1. The cylindrical bucket (2.540 cm o.d., 1.925 cm i.d., and 8.25 cm deep) is a composite of stain-

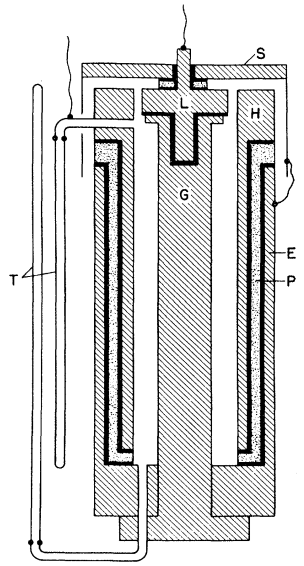

FIG. 1. Schematic drawing of bucket capacitor.