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## ATTEMPTS TO DETECT RESONANCE SCATTERING IN $Zn^{67}$ ; THE EFFECT OF ZERO-POINT VIBRATIONS\*

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The 93-keV  $\gamma$  ray from the 9.4- $\mu$ sec level of  $Zn^{67}$  would appear to offer an extreme example of the fractionally narrow  $\gamma$  rays in solids discovered by Mössbauer.<sup>1</sup> It has been suggested as an ideal medium for studying such small frequency shifts as would result from, for example, gravity.<sup>2,3</sup> We wish to describe some unsuccessful attempts to detect resonant absorption of this  $\gamma$  ray and to offer an explanation which has more general applicability and complicates the extension of the technique much beyond the narrowness presently achievable with  $Fe^{57}$ .<sup>4-7</sup>

The 93-keV  $\gamma$  ray is emitted as the final step in the decay of 78-hr  $Ga^{67}$  to stable  $Zn^{67}$ .  $Ga^{67}$  was prepared with good radiochemical purity by ( $\alpha$ ,  $2n$ ) reaction in  $Cu^{65}$  by about 10 microampere hours of bombardment by the MIT cyclotron.

Resonant absorption experiments were attempted for one combination of source and absorber at the temperature of liquid nitrogen and, for this and three other combinations, at the temperature of liquid helium. The experiments at helium temperature used a cryostat with a thin bottom to allow the 93-keV  $\gamma$  ray to be measured externally. In all experiments comparison was made of the  $\gamma$ -ray intensity transmitted through the absorber with the source stationary to that with it vibrating at an rms velocity of about  $10^{-3}$  cm/sec, about one hundred times larger than the expected linewidth. The vibration was produced by a quartz electromechanical transducer which was also cooled. In all cases a null effect was found to the accuracy of the statistical uncertainty of  $\pm 0.1\%$

of the counts in one channel. Correcting for background with the tenfold nonresonant absorption normally present in the absorbers leaves the result that no resonant absorption greater than 0.3% was present in any of the combinations. The combinations tried were:

1. Source  $Ga^{67}$  dissolved in 95% aluminum 5% by weight zinc alloy, and absorber 90% aluminum 10% zinc by weight, 4 cm thick. It was hoped that the aluminum would provide a cubic environment to avoid electric quadrupole splitting as well as a moderately high effective Debye temperature. Unfortunately the necessarily high zinc concentration probably destroys lattice symmetry seriously enough to broaden the lines through the inhomogeneous electric quadrupole interaction. The 11-kc/sec linewidth in the absorber resulting from the 9.4- $\mu$ sec half-life is comparable to widths obtained in nuclear paramagnetic resonance and it is known that even 0.5% of zinc broadens the Al magnetic resonance line.<sup>8</sup> One would not expect the Zn to be less affected. Ignoring that broadening and supposing, optimistically, that the 375°K Debye temperature of the Al host applied to the recoil dynamics of the zinc, one would expect an effect of 0.4% at nitrogen temperature and 2.2% at helium temperature.

2. Source  $Ga^{67}$  dissolved in polycrystalline zinc and absorber 5-mm thickness of polycrystalline zinc. In this case one would expect a reduction of the cross section by a factor of 13/50 because of electric quadrupole splitting in the hexagonal close-packed lattice. The Debye tem-

perature of zinc is strongly temperature dependent. If one uses a value of 240°K, one expects only 0.08% effect, but if one uses the value of 305°K characteristic of the specific heat at 4°K or of the elastic constants, one expects 0.56%. Undoubtedly this estimate is overly optimistic because the low-temperature specific heat measurements and the elastic constants relate only to the low-frequency end of the spectrum of lattice waves whereas the high-frequency part dominates the recoil dynamics.

3. Source Ga<sup>67</sup> dissolved into a single crystal of zinc with a 5-mm-thick single zinc crystal absorber. These crystals were oriented with *c* axes perpendicular to the direction of propagation of the  $\gamma$  rays observed and parallel to one another. It was hoped that the relative hardness of zinc in the direction perpendicular to the *c* axis would enhance the recoil-free component of the  $\gamma$  ray above that predicted from the Debye temperature.<sup>9</sup> Again, this is mainly connected with the low-frequency part of the phonon spectrum. In addition, the quadrupole splitting would reduce the cross section less with oriented single crystals than with polycrystalline samples.

4. Source Ga<sup>67</sup> left in the cubic copper foil but annealed to relieve radiation damage, and about one-centimeter thickness of natural sphalerite, ZnS, from Franklin, New Jersey, reputedly of relatively high purity, used as absorber. In this case broadening and reduction of cross section by electric quadrupole coupling should have been avoided if the sphalerite was adequately pure. The moderately high Debye temperature of Cu should prove helpful. Using 315°K for the Debye temperature of copper and 270°K for ZnS leads to an expected effect of 1.15%.

The failure of the above attempts may involve several causes. The effects of the electron capture that converts Ga<sup>67</sup> to Zn<sup>67</sup> and of the  $\gamma$  rays leading to the 9.4- $\mu$ sec state may not be healed adequately quickly and completely at helium temperatures to avoid broadening by electric quadrupole interactions. More fundamental, however, is an effect related to that involved in the predicted<sup>10, 11</sup> and observed<sup>10</sup> variation with temperature of the frequency of the  $\gamma$  ray of Fe<sup>57</sup>. The fractional width of the unbroadened line in Zn<sup>67</sup> should be only 10<sup>-15</sup>. It is now clear that the lattice energy shifts the frequency of the  $\gamma$  ray through the second order Doppler effect and that when different lattices are used for source and absorber the  $\gamma$ -ray and absorption line will be at different frequencies. Large shifts compared to

10<sup>-15</sup> have been found between source and absorbers of Fe<sup>57</sup> even when both are at a common temperature and ostensibly the same material.<sup>12</sup> Differences of Debye temperature were suggested as the cause. Differences of Debye temperature should result in such relative shifts even at the absolute zero of temperature owing to the difference in zero-point energies. Assuming a Debye spectrum for the lattice oscillators, the zero-point energy of  $9k\theta_D/8$  per atom leads for Zn<sup>67</sup> to a fractional decrease of frequency of 0.772  $\times 10^{-15}$  per degree increase in Debye temperature. If the zero-point vibration of the Zn<sup>67</sup> in copper has an effective Debye temperature 45°K higher than that of the Zn<sup>67</sup> in ZnS, then the absorption line in the ZnS would be fractionally 35 full line-widths higher in frequency.

Resonant absorption could be observed only by compensating that shift by a constant relative velocity. To find the line without prior knowledge of the displacement would require an extensive search and a good velocity spectrometer. Furthermore, inhomogeneity of the lattice could introduce important broadening and consequent weakening of the lines through small variations of the zero-point energies. Only in the experiments using zinc directly, of those reported above, would there appear to have been hope of finding absorption by the method used. For those cases the effect expected was probably too small to be observed.

Addition of the effect of the zero-point vibrations to the shifts induced thermally modifies the values of the differences in  $\theta_D$  needed to explain the shifts at a common temperature between absorber and source reported previously for Fe<sup>57</sup>.<sup>12</sup>

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<sup>1</sup>R. L. Mössbauer, *Z. Physik* **151**, 124 (1958); *Naturwissenschaften* **45**, 538 (1958); *Z. Naturforsch.* **14a**, 211 (1959).

<sup>2</sup>R. V. Pound and G. A. Rebka, Jr., *Phys. Rev. Letters* **3**, 439 (1959).

<sup>3</sup>I. Ya. Barit, M. I. Podgoretsky, and F. L. Shapiro, *Zhur. Eksp. i Teoret. Fiz.* **38**, 301 (1960).

<sup>4</sup>R. V. Pound and G. A. Rebka, Jr., *Phys. Rev. Letters* **3**, 554 (1959).

<sup>5</sup>J. P. Schiffer and W. Marshall, *Phys. Rev. Letters* **3**, 556 (1959).

<sup>6</sup>S. S. Hanna, J. Heberle, C. Littlejohn, G. J. Perlow, R. S. Preston, and D. H. Vincent, *Phys. Rev. Letters* **4**, 28 (1960).

<sup>7</sup>G. De Pasquali, H. Frauenfelder, S. Margulies, and R. N. Peacock, *Phys. Rev. Letters* **4**, 71 (1960).

<sup>8</sup>T. J. Rowland, *Acta Met.* **3**, 74 (1955).

<sup>9</sup>We wish to thank Professor Bruce Chalmers for pointing out the large anisotropy of elastic constants

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<sup>10</sup>R. V. Pound and G. A. Rebka, Jr., *Phys. Rev. Letters* **4**, 274 (1960).

<sup>11</sup>B. D. Josephson, *Phys. Rev. Letters* **4**, 341 (1959).

<sup>12</sup>R. V. Pound and G. A. Rebka, Jr., *Phys. Rev. Letters* **4**, 337 (1960).

### SEARCH FOR THE ANISOTROPY OF INERTIA USING THE MÖSSBAUER EFFECT IN Fe<sup>57</sup>†

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The width of the central resonance absorption line in the Mössbauer effect in Fe<sup>57</sup> has been found by several observers to be considerably greater than the natural linewidth.<sup>1-4</sup> In all these cases the emitting and the absorbing nuclei were randomly oriented, and the possibility arises that this widening is due to an external perturbation which affects the nuclear levels.

An intriguing mechanism for line broadening has been pointed out by Cocconi and Salpeter.<sup>5,6</sup> The locally asymmetric distribution of matter in the universe, in accordance with Mach's principle, may cause a local anisotropy  $\Delta M = \frac{2}{3}(M' - M'')$  in inertia. Here  $M'$  is the inertial mass for matter accelerated towards the center of the Galaxy and  $M''$  the inertial mass for acceleration perpendicular to this direction. Such an anisotropy  $\Delta M$  would give rise to a shift in atomic<sup>5</sup> and nuclear<sup>6</sup> energy levels. In the case of Fe<sup>57</sup>, the shift would occur only for the excited state with spin 3/2. Cocconi and Salpeter compute the level shift  $\Delta E$  to be  $(\Delta M/M)\bar{T}\bar{P}_2$ , where  $\bar{T}$  is the average kinetic energy of the nucleon responsible for the transition, and  $\bar{P}_2$  has the value 1/5 for the most favorable relative orientation between the nuclear angular momentum and the line to the galactic center. Assuming  $\bar{T} = 10$  Mev and a line broadening of  $10^{-8}$  ev, Cocconi and Salpeter conclude that  $\Delta M/M$  has an upper limit of about 1 part in  $10^{14}$ .

Since the excess broadening of the center resonance line is large, it is easy to determine whether it arises from effects due to the Galaxy. In our first test, we placed both the Fe<sup>57</sup> source and the Fe absorber in parallel magnetic fields of about 1000 gauss. In this condition, the atomic magnetic fields at the emitting and absorbing nuclei are parallel and the postulated galactic

shifts should be the same for each of the corresponding magnetic sublevels in source and absorber. Thus any line broadening due to anisotropic inertia should disappear. The experiment showed, however, that over a twenty-four hour period there was no significant change in the linewidth between the two conditions: (1) no magnetic field, random domain alignments, and (2) source and absorber in parallel magnetic fields. The accuracy of this test is limited by the fact that, although the domains are aligned, the local magnetic fields at the nuclei in the source and absorber are different, producing an additional broadening effect. The results showed clearly that the majority of the excess broadening was not due to the Galaxy.

To perform a more sensitive test for a possible galactic effect, a magnetic field of about 600 gauss at the moving horizontal absorber<sup>3</sup> was aligned in the north-south direction. The source was placed in a magnetic field of similar magnitude, but lying in a vertical, north-south plane, and oriented at +45° with respect to the horizontal (see Fig. 1). Each sidereal day, the galactic center rotates once around a line parallel to the earth's axis, changing its direction with respect to the magnetic fields of source and absorber. Cocconi and Salpeter assume that the level shift reaches an extreme value when the magnetic field  $H$  is perpendicular to the line toward the galactic center, and again when it is parallel to that direction. At an intermediate angle the level shift vanishes.

In the experimental arrangement, shown schematically in Fig. 1, there is an interval of about 10 hours, centered at 17:30 sidereal time, when the line toward the galactic center makes nearly equal angles with the two magnetic field direc-