# THE MÖSSBAUER EFFECT\*

#### F. L. SHAPIRO

#### Usp. Fiz. Nauk 72, 685-696 (December, 1960)

# 1. CLASSICAL THEORY OF THE MÖSSBAUER EFFECT

As Mössbauer<sup>1</sup> has shown, the spectrum of  $\gamma$  rays emitted by excited nuclei which are contained in a solid consists of two components:

a) a component with the natural width and with an energy equal to the transition energy E;

b) a much wider component with a width of the order of Ev/c (v is the mean square velocity of thermal motion of the radiating nucleus), shifted with respect to the transition energy by the amount R, where R =  $E^2/2Mc^2$  is the recoil energy in the emission of a quantum by the free nucleus with mass M, at rest.

The same components are also present in the absorption spectrum by the corresponding unexcited nucleus. The component with the natural width we shall in the following call the Mössbauer line.

In quantum theory<sup>1</sup> the Mössbauer line corresponds to acts of radiation (or absorption) of  $\gamma$  quanta which occur elastically with respect to the crystal as a whole, i.e., without any change in the quantum state of the crystal, or, in other words, without radiation or absorption of phonons. However, it turns out that the existence of the Mössbauer line is a purely classical effect which can be described quantitatively also without the use of quantum theory.<sup>2</sup>

From the classical point of view the thermal vibrations of the radiating atom produce a phase or, what is equivalent, a frequency modulation of the  $\gamma$  radiation via the Doppler effect. A large number of satellite lines appear then in the spectrum of the radiation, in addition to which there is also always the central unshifted component (the carrier frequency), which is the Mössbauer line. Let us find the intensity of this component.

Let u(t) be the projection of the velocity of the radiating atom on the direction of observation. If we include the linear Doppler effect, the phase of the wave in the laboratory coordinate system is

$$\Psi(t) = \int \omega_0 \left(1 + \frac{u(t)}{c}\right) dt = \omega_0 t + \frac{x(t)}{\lambda},$$

where  $\omega_0$  is the frequency of the  $\gamma$  radiation,  $\pi = c/\omega_0$ is the wave number, x(t) is the displacement of the atom from its equilibrium position along the direction of observation.

Expanding x(t) in series in the natural frequencies

of oscillation of the solid, we have for the field intensity in the wave,

$$\mathscr{E} = e^{i\psi(t)} = e^{i\omega_0 t + i \sum_{m=\pi}^{\infty} \frac{\Sigma^{m} \sin(\Omega_m t + \varphi_m)}{m \, \pi}}$$

Using the familiar expansion from the theory of Bessel functions,

$$e^{iz\,\sin\theta} = \sum_{n=-\infty}^{\infty} J_n(z)\,e^{in\theta},$$

we obtain

$$\mathscr{E} = e^{i\omega_0 t} \prod_m \sum_{n=-\infty}^{\infty} J_n\left(\frac{x_m}{\lambda}\right) e^{in\left(\Omega_m t + \varphi_m\right)}$$
(1)

The general term in (1) corresponds to radiation of frequency  $\omega_0 + \sum_m n_m \Omega_m$ . The unshifted frequency is given by the term  $\mathscr{E}_0 = e^{i\omega_0 t} \prod_m J_0(x_m/\pi)$ , and also by those terms for which  $\sum_m n_m \Omega_m = 0$ . In the cases of practical interest, when the intensity of the Mössbauer line is not too small, the terms of the latter type can be neglected since the number of such terms is small compared with the total number of satellite lines.

Thus the intensity of the Mössbauer line is equal to

$$f = |\mathscr{E}_0^2| = \prod_m J_0^2 (x_m/\lambda_m).$$

Since the number of factors is very large, each of the quantities  $J_0(x_m/\lambda)$  is close to 1, and we may set  $J_0(x_m/\lambda) = 1 - x_m^2/4\lambda^2$ . As a result we get

$$\ln t = -\frac{1}{2\lambda^2} \sum_m x_m^2 = -\frac{\overline{x^2}}{\lambda^2}, \qquad (2)$$

where  $\overline{x^2} = \frac{1}{2} \sum x_m^2$  is the mean square deviation of the vibrating atom in the lattice from its equilibrium position (along the direction of observation).

From (2) it follows that the Mössbauer line is well developed if the amplitude of oscillation of the atoms of the lattice is not large compared to the wavelength of the  $\gamma$  radiation. The meaning of this condition can be understood if we use an approach applied by I. M. Frank in the theory of the Doppler effect.<sup>3</sup> The radiation from an atom can be regarded as the result of a superposition of light pulses emitted during successive intervals dt<sub>i</sub>. The spectral distribution of the individual pulse corresponds to a continuous spectrum, i.e., it contains all frequencies. For a radiator at rest the phase relations in the spectral distribution are such that waves with frequencies  $\omega \neq \omega_0$  cancel one another, whereas the waves with frequency  $\omega_0$  reinforce.

<sup>\*</sup>This is the revised text of a report presented to the Second All-Union Conference on Nuclear Reactions at Low and Medium Energies (July, 1960).

If the radiator vibrates with an amplitude small compared to the wavelength of the radiation  $\lambda$ , i.e., x  $\ll \lambda$ , the phase shifts are changed insignificantly, and the intensity of the component with frequency  $\omega_0$  also is reduced insignificantly. However, if  $x > \lambda/2$ , the waves with frequency  $\omega_0$  begin to cancel one another, and the intensity of the component  $\omega_0$  falls sharply.

For an oscillator with natural frequency  $\Omega$ , the mean square displacement and the energy U are related by the familiar equation

$$\bar{x^2} = \frac{U}{M\Omega^2} ; \qquad (3)$$

The mean energy of an oscillator in turn is given by the expression

$$\widetilde{U} = \hbar \Omega \left( \frac{1}{2} + \frac{1}{e^{\hbar \Omega/kT} - 1} \right).$$
(4)

Combining (3) and (4) and, after averaging over the spectrum of vibrations of the solid, substituting the result in (2), we get

$$\ln f = -\frac{E^2}{Mc^2} \int_0^\infty \left[ \frac{1}{2} + \frac{1}{e^{\hbar\Omega/kT} - 1} \right] \frac{f(\Omega) \, d\Omega}{\hbar\Omega} , \qquad (5)$$

where T is the absolute temperature,  $f(\Omega)$  is the spectrum of the natural frequencies of the solid normalized to unity.

In the Debye approximation,

$$f(\Omega) = 3\left(\frac{\hbar}{k}\right)^3 \frac{\Omega^2}{\theta^3} \text{ for } \hbar\Omega \ll k\theta, \ f(\Omega) = 0 \text{ for } \hbar\Omega > k\theta$$

( $\theta$  is the Debye temperature). Substituting this expression in (5), we arrive at formula (1) of Mössbauer's paper (cf. earlier on page 867). Thus the results of the classical and quantum calculations are in complete agreement.

The expression for f is essentially a special case of the so-called Debye-Waller temperature factor which takes into account the influence of the vibrations of the atoms in the lattice on the intensity of the coherent Bragg scattering of x-rays.<sup>4</sup> This identity of the results reflects a common feature of the two phenomena: both the Mössbauer effect and Bragg scattering are given by the undisplaced component in the frequency spectrum of the radiation, modulated by the vibrations of the lattice atoms; in the first case we deal with vibrations of a radiating atom, and in the second with those of a scattering atom.

As is well known, many materials, especially those not having cubic symmetry, cannot be characterized by a definite Debye temperature.<sup>5</sup> This results in a considerable uncertainty in estimating the intensity of the Mössbauer line from relations obtained for the Debye model. M. V. Kazarnovskii has succeeded in expressing the intensity of the Mössbauer line directly in terms of an integral of the lattice specific heat  $C_L$ over the temperature.<sup>6</sup> Thus for T = 0 (or  $T \ll \theta$ )

$$\ln f = -\frac{E^2}{2\pi^2 M c^2 k^2} \int_0^\infty C_L(T) \frac{dT}{T^2}.$$
 (6)

Kazarnovskii's relations are very useful for comparing experiment with theory. Unfortunately, they are rigorously valid only for monatomic crystals with cubic symmetry, for which they are less necessary.

# 2. THE INFLUENCE OF THE QUADRATIC DOPPLER EFFECT

Pound and Rebka<sup>7</sup> discovered the dependence of the frequency of the Mössbauer line on the temperature of the source (or absorber) and showed that it is due to a Doppler effect of second order in the velocity of the thermal motion, which we neglected earlier.

The exact relativistic formula for the Doppler effect has the form

$$\omega = \omega_0 \frac{\sqrt[V]{1-(v/c)^2}}{1-u/c} ,$$

where the velocity of the radiator, v, and the projection of the velocity on the direction of observation in the lab coordinate system, u, must be taken at the instant of radiation  $\tau$ , which is related to the moment of observation t by the relation

$$t = \tau + \frac{X - x(\tau)}{c} . \tag{7}$$

Here X is the distance of the point of observation from the equilibrium position of the radiator (it is assumed that this distance is infinitely large compared to the amplitude of vibration of the radiator).

The frequency  $\omega_1$  of the Mössbauer line is equal to the constant term in  $\omega(t)$ , whereas the variable term in  $\omega(t)$  determines, as we have shown above, its intensity. Thus

$$\omega_{1} = \frac{1}{T} \int_{0}^{T} \omega(t) dt = \frac{1}{T} \int_{\tau_{1}}^{\tau_{2}} \omega_{0} \frac{\sqrt{\frac{t}{1 - v^{2}}(\tau)/c^{2}}}{1 - u(\tau)/c} \frac{dt}{d\tau} d\tau \simeq \omega_{0} \left(1 - \frac{\overline{v^{2}}}{2c^{2}}\right),$$
(8)

since according to (7),  $dt/d\tau = 1 - u(\tau)/c$  and (for an atom in a solid)  $(\tau_2 - \tau_1)/T \rightarrow 1$  for  $T \rightarrow \infty$ .

Equation (8) simply expresses the relativistic "clock paradox." A moving clock runs slower than the clock of the observer by the factor  $\sqrt{1-v^2/c^2}$   $\simeq 1-v^2/2c^2$ . It can also be obtained from energy considerations.<sup>2,8</sup> In fact, in the Mössbauer effect the recoil momentum is transmitted to the crystal as a whole, i.e., the momentum **p** of the radiating atom is not changed. But, since the mass of the radiating atom decreases by the amount  $\Delta M = E/c^2$ , its average kinetic energy  $\overline{p^2}/2M$  increases by an amount  $\frac{\overline{p^2}\Delta M}{2M^2} = E \frac{\overline{v^2}}{2c^2}$ . Corresponding to this, the energy of the radiated  $\gamma$  quantum turns out to be less than the transition energy E by a factor  $(1-\overline{v^2}/2c^2)$ .

Differentiating (9) with respect to the temperature and using the fact that the heat capacity of the lattice  $C_{\rm L} = du/dT$ ,  $u = M\overline{v^2}$ , we obtain the relation of Pound and Rebka

$$\frac{d\omega_1}{dT} = -\omega_0 \frac{C_L}{2Mc^2} \,. \tag{9}$$

In the preceding treatment we have assumed that the vibrations do not affect the frequency of the  $\gamma$  radiation in the coordinate system of the radiating nucleus. Actually, the displacement of an atom from its equilibrium position results in a change in the configuration of the electron shells and consequently in a change in the interaction energy of the shell with the nucleus.\* This change is different for the excited and ground states of the nucleus, and this difference enters into the energy of the  $\gamma$  quantum. This question has been treated in detail in a paper by Dzyub and Lubchenko.<sup>9</sup>

The order of magnitude of this difference can be estimated from the available data on isomeric shifts, i.e., on the shift in energy of atomic optical transitions for the ground and isomeric states of a nucleus.<sup>10</sup> In order of magnitude, the isomeric shift amounts to  $E_{isom} \sim 10^{-6}$  ev. The relative change in isomeric shift can be set equal to the relative change in energy of the electronic transition, i.e., to the width of the optical line in the crystal:

$$\Delta E_{isom} \sim \frac{\Delta E_{opt}}{E_{opt}} E_{isom} \sim 10^{-3} \cdot 10^{-6} \sim 10^{-9} \text{ ev.}$$

Since when a sample is heated the amplitude of the displacements increases, the observed effect should lead to a temperature dependence of the position of the resonance line. The good agreement of the experimental data<sup>7</sup> for  $Fe^{57}$  with the theory, when one takes into account only the quadratic Doppler effect, shows that for  $Fe^{57}$  the change in the isomeric shift is not significant.<sup>†</sup>

# 3. MÖSSBAUER EFFECT FOR AN IMPURITY

The predominant part of the normal vibrations of a lattice is characterized by a wavelength considerably greater than the dimensions of the elementary cell of the lattice. For such vibrations the displacements of the atom with respect to its nearest neighbors are small compared to the amplitude of the displacement with respect to its equilibrium position, i.e., a large number of atoms practically vibrate together. Because of this, the replacement of one of the atoms in the lattice (mass  $M_1$ ) by another atom with mass M affects the dynamics of the oscillations only insignificantly, and the displacement of the impurity atom will differ little from the displacement of the atoms of the lattice. The only exceptions are the few high frequency oscillations associated with sizable displacements of the atom M with respect to its

neighbors. These few high frequency vibrations make a very small contribution to the mean square displacement  $\overline{x^2}$ , especially since for the individual oscillator  $\overline{x_m^2} \sim 1/\Omega_m$ . Because of this, the intensity of the Mössbauer line in the radiation of the impurity atom should be almost the same as it would be if a  $\gamma$  ray of the same wavelength were radiated by the fundamental component atoms of the lattice. This means that in the case of an impurity atom we should substitute in expression (5) the mass of the atom and the vibration spectrum characterizing the lattice in which the impurity atom has been introduced. For a Debye crystal with Debye temperature  $\theta$  and for  $T \ll \theta$  we shall have for the radiation by the atom M

$$\ln f \simeq -\frac{3E^2}{4M_1c^2k\theta} , \qquad (10)$$

whereas for a crystal with the same Debye temperature consisting entirely of atoms M, we have

$$\ln f = -\frac{3E^2}{4Mc^2k\theta} . \tag{11}$$

As we see from (10), in the case of the impurity atom, the intensity of the Mössbauer line is no longer determined by the recoil of the radiating atom.

The remarks made above have still not been checked by experiment or exact computation. They may have practical significance for observation of the Mössbauer effect on nuclei of low and medium mass numbers.<sup>11</sup> For example, in metallic zinc ( $\theta = 213^{\circ}$ K) the intensity of the Mössbauer line of Zn<sup>67</sup> should amount to f = 0.4% for  $T \ll \theta$ . If we introduce Zn<sup>67</sup> as an impurity in a lattice with the same Debye temperature, but with a mass number for its atoms amounting to 200, the quantity f should, according to (10), increase by a factor of approximately 40. Here one should remember that the frequency of the resonance radiation of an atom depends on its crystalline and chemical surroundings [cf. Eq. (8) above and also reference 12].

In the case of a complex lattice containing atoms of more than one type, the atoms in the cell move practically together during vibrations of the lattice, except for the high-frequency part of the optical branch of the vibration spectrum, where the displacements of the atoms in a cell are different. If we neglect this last fact, then, as is easily shown, in calculating the intensity of the Mössbauer line we must substitute a mass which is an average of the masses of all the atoms in a cell. However, the admissibility of such an approximation is by no means evident, and the question requires more detailed investigation.

#### 4. COHERENT EFFECTS

In a paper<sup>13</sup> by Podgoretskiĭ and Roĭzen they point out that the Mössbauer effect makes possible the migration of excitation between identical nuclei; this influences the character of the radiation. In the paper they consider classically the cases of a diatomic mole-

<sup>\*</sup>Changes in energy of the nucleus as a result of a varying polarization, under the action of the forces produced by the oscillation, are negligibly small.

<sup>&</sup>lt;sup>†</sup>The frequency change  $\Delta E_{isom}/\hbar$  leads to a phase shift of the radiated wave by an amount  $-\Delta E_{isom}T/\hbar$ , where  $T \sim \hbar/k\theta$  is the period of vibrations of the solid. This shift is negligibly small compared to unity, i.e., it has practically no effect on the intensity of the Mössbauer line.

cule and a long linear chain of atoms. In the case of the chain, the frequency of the radiation and the damping constant depend on the angle between the direction of the ray and the axis of the chain. The frequency displacement, measured in natural line widths, is equal to  $3\lambda/16\pi a$  (except for certain special angles); the relative change in the damping (as compared to the free nucleus) amounts to  $3\lambda/8a$ , where  $\lambda$  is the wavelength and a is the distance between atoms in the chain. The authors point out that analogous phenomena should be observed in crystals. These effects can reach their maximum magnitude only in monoisotopic substances and for an intensity of the Mössbauer line close to unity.

### 5. SPECTRUM OF GAMMA RADIATION FROM GASEOUS AND LIQUID SOURCES

Since in a liquid or a compressed gas the displacement of a diffusing atom during the lifetime of the excited nucleus is small, the question arises whether a sharp  $\gamma$  line can also be formed in such media. The corresponding problem was considered (essentially classically) in a paper of Podgoretskiĭ and Stepanov.<sup>14</sup> These authors, using the Langevin equation for describing the motion of a diffusing atom, obtained the following expression for the shape of the emission line:

$$I(\omega) \sim \operatorname{Re} \int_{0}^{\infty} dt e^{-i(\omega-\omega_{0})t-\gamma t/2-(\eta t-1+e^{-\eta t})D/\eta \lambda^{2}}, \qquad (12)$$

where  $\omega_0$  is the frequency emitted by a fixed nucleus,  $\gamma = \Gamma/\hbar$  is the decay constant of the excited state, D is the diffusion coefficient, and  $\eta = kT/MD$ .

For  $D/\eta \lambda^2 \sim (l/\lambda)^2 \gg 1$  (where *l* is the mean free path between collisions) only small values of  $\eta t$ are important in (12), and the last term in the exponential in (12) takes the form  $D\eta t^2/2\lambda^2$ . The spectrum in this case has a Doppler shift with a half-width of the order of  $\omega_0 v/c = v/\lambda$ , where v is the mean square velocity of the thermal motion. (It is assumed that  $\gamma \ll \omega_0 v/c$ .) In the opposite case, when  $D/\eta \lambda^2 \sim (l/\lambda)^2$  $\ll 1$ , large values of  $\eta t$  are significant in (12), and the last term in the exponent can be set equal to  $Dt/\lambda^2$ ; the line takes on the Lorentz shape,

$$I(\omega) \sim 1/[(\omega - \omega_0)^2 + (\gamma/2 + D/\lambda^2)^2]$$
 (13)

with a width  $\gamma/2 + D/\lambda^2$ .

Since  $D/\lambda^2 \sim \frac{v}{\lambda} \cdot \frac{l}{\lambda}$ , we see that in the region of

applicability of formula (13) the width of the spectrum is much less than the Doppler width  $v/\lambda$ . For  $\gamma$  radiation with an energy of 10 kev and for a diffusion coefficient of  $10^{-5}$  cm<sup>2</sup>/sec (liquid lead), the additional width is equal to  $\hbar D/\lambda^2 \sim 10^{-3}$  ev, i.e., large compared with the natural width of the  $\gamma$  line.

Lipkin<sup>15</sup> obtained a sum rule according to which the average energy of the emission line is always less than the energy of the  $\gamma$  transition by the amount of the re-

coil energy R. In a solid when there is an unshifted line the fulfillment of the sum rule is accomplished by a shift in the Doppler component of the spectrum. According to the classical theory, for a liquid or a gas the spectrum of the radiation consists of a single line whose center coincides with the frequency of the transition. For  $E \rightarrow 0$  the ratio of the recoil energy to the line width is constant and equal to  $\hbar/2MD$ . This quantity may not be small. The question as to how the sum rule is satisfied and, consequently, the question of the location of the center of the line cannot be answered completely in classical theory and remains unclear at present.

### 6. EXPERIMENTS WITH Sn<sup>119</sup>

Tin-119 has an excited state with an excitation energy of 23.8 kev and a width  $\Gamma = 2.4 \times 10^{-8}$  ev. The spin of the ground state of  $\operatorname{Sn}^{119}$  is  $I_0 = \frac{1}{2}^+$ ; for the excited state  $I_1 = \frac{3}{2}$ . This level is excited as a result of decay of the isomeric state of  $\operatorname{Sn}^{119}$  with a half-life of 250 days.

The Mössbauer effect in tin-119 has been investigated by a group at the Institute for Theoretical and Experimental Physics (ITÉF) (Alikhanov and Lyubimov<sup>16,17</sup>), the Physics Institute of the Academy of Sciences (Barit et al.<sup>18</sup>), and Moscow State University (Delyagin, Shpinel' et al.<sup>19,20</sup>). The work of the group at the ITÉF is the first in which the resonance absorption effect in tin was observed and the influence of a magnetic field on the absorption was measured. The measurements were carried out at liquid-nitrogen and at room temperatures. The source and filter were prepared from metallic white tin. The results of the measurements in which the source was placed in a magnetic field and the absorber of natural tin 5 mg/  $cm^2$  thick was outside the field are shown in Fig. 1.



FIG. 1. Dependence of the magnitude of resonance absorption in a thin sample of Sn<sup>119</sup> on the intensity of the magnetic field acting on the source<sup>17</sup> (H in oersted). The magnitude of the resonance absorption was determined as the difference in intensity of the transmitted radiation for temperatures of source and filter equal to 290 and 90° K. The solid curves were calculated on the assumptions: 1)  $\mu_1 = -0.25$  nuclear magnetons [no quadrupole splitting ( $\Delta = 0$ )], 2)  $\mu_1 = 0$ ,  $\Delta = \Gamma/2$ , 3)  $\mu_1 = 1.0$  nuclear magnetons,  $\Delta = 0.95\Gamma$ .

The reduction of the resonance absorption when the field is applied results from the splitting of the level as a result of a nuclear Zeeman effect, whose magnitude is determined by the magnetic moments of the ground and excited states of Sn<sup>119</sup>. The magnetic moment of the ground state of  $\operatorname{Sn}^{119}$  is known<sup>21</sup> ( $\mu = -1.05$ nuclear magnetons). In principle, from the curve of Fig. 1 one could obtain the value of the magnetic moment of the excited state of Sn<sup>119</sup>. Such a deduction is made difficult by the need to take into account a) the actual width of the resonance line, which frequently is greater than the natural width, and b) the splitting of the excited state of  $\operatorname{Sn}^{119}$  (spin  $\frac{3}{2}$ ) because of electric quadrupole interaction with the inhomogeneous electric field of the hexagonal lattice of tin.

According to the computations of Alikhanov and Lyubimov,<sup>17</sup> broadening of the line to double the natural width changes the sign of the magnetic moment of the excited level from -0.25 to +1.0 nuclear magnetons.

In the presence of a quadrupole splitting which is large compared to the Zeeman splitting, the size of the latter depends on the angle between the direction of the magnetic field and the symmetry axis of the electric field. For a level with spin  $I = \frac{3}{2}$ , for the components with projection of the angular momentum equal to  $\pm \frac{3}{2}$ , the interaction energy is equal to<sup>22</sup>

$$W = \frac{1}{2}\Delta \pm \mu H |\cos\theta|. \tag{14}$$

For the components  $\pm \frac{1}{2}$ ,

$$W = -\frac{1}{2}\Delta \pm \frac{2}{3}\mu H \sqrt{1 - \frac{3}{4}\cos^2\theta} .$$
 (15)

From expressions (14) and (15) it follows that in a polycrystalline sample the presence of a quadrupole splitting changes the mean value of the magnetic splitting and leads to a broadening of the Zeeman components.

Since at present there are no data concerning the presence or absence of broadening of the resonance line of  $\text{Sn}^{119}$ , and since the quadrupole splitting has still not been taken into account, we may speak only concerning a preliminary value for the magnetic moment of the excited level of  $\text{Sn}^{119}$ . As such a value, the authors give  $\mu_1 = -0.25$  n.m.

More simple in their interpretation are experiments in which one observes the shape of the resonance curve. Such experiments were carried out by the group at Moscow State University. The measurements were made at liquid nitrogen temperature. By using a shaped cam, the source was given a velocity which varied linearly with time over a definite range. The pulses from the  $\gamma$  ray detector entered a circuit which modulated the pulse amplitudes linearly in synchronism with the change in velocity of the source, and then the pulses went to a 100-channel pulse-height analyzer. Such an arrangement enables one to meas-

ure simultaneously the whole absorption spectrum of the  $\gamma$  rays as a function of the relative velocity. The results of the measurements<sup>19</sup> with the source in the form of metallic white tin and filters of white tin and of SnNb<sub>3</sub> are shown in Fig. 2. These data show the presence of a quadrupole splitting in white tin and the absence of any significant splitting in the SnNb<sub>3</sub> alloy. For the magnitude of the separation between the components of the quadrupole splitting one gets the value  $\Delta = (1.15 \pm 0.25) \times 10^{-7}$  ev.

In Fig. 3 are shown the results of measurements in which the absorber, an SnNb<sub>3</sub> alloy, was placed in a magnetic field of 12,000 oersted, whereas the source of metallic tin was not in the field.<sup>20</sup> One observes a very definite splitting of the line corresponding to the picture computed for a value of  $\mu_1 = (1.9 \pm 0.4)$  n.m. However, the precision of the measurements is low.



FIG. 2. Dependence of intensity of the 23.8-kev  $\gamma$  radiation of tin, passing through filters of tin and SnNb<sub>3</sub> alloy, on the relative velocity of source and filter.<sup>19</sup> (The abscissa is v, mm/sec.)



FIG. 3. Influence of magnetic field on intensity of filtered radiation<sup>20</sup> from the 23.8-kev level of Sn<sup>119</sup>. The source was metallic tin, the filter an alloy of SnNb<sub>3</sub>. Below is shown the pattern expected for a very narrow line on the assumption that  $\mu_0 = 1.05$  n.m.,  $\mu_1 = 1.9$  n.m.

To check the agreement of experiment with theory, the group at the Physics Institute of the Academy of Sciences measured resonance absorption at various temperatures and for various absorber thicknesses.<sup>18</sup> The results for liquid nitrogen temperature are shown in Fig. 4. The theoretical curves were constructed taking into account the quadrupole splitting according to the data of the Moscow State University group and on the assumption that there is no broadening of the line. These results and data on measurements at higher temperatures are in agreement with an effective Debye temper-



FIG. 4. Dependence of magnitude of resonance absorption of  $\gamma$  rays of Sn<sup>119</sup> on thickness of filter (metallic tin).<sup>18</sup> G =(I<sub>0</sub> - I)/I<sub>0</sub> is the relative magnitude of the resonance absorption; x = nf  $\sigma_0$ , where n is the number of atoms of Sn<sup>119</sup> per square centimeter of the filter, f' is the intensity of the Mössbauer line for the filter,  $\sigma_0$  is the cross section at resonance. The solid curves are theoretical curves computed including quadrupole splitting;  $\alpha$  is the fraction of the radiation of the 23.8-kev line of Sn<sup>119</sup> in the recorded  $\gamma$  intensity of the source.

ature for tin ~180°K. Preliminary data from measurements with a moving source confirm the conclusion of the Moscow State University group concerning the presence of a quadrupole splitting. However, the quadrople splitting did not appear in the work of reference 23, which was carried out in France. Since the experiments at Moscow State University definitely show the presence of quadrupole splitting, one may surmise that the discrepancy is associated with an error in the velocity scale in one of the two papers, or with some apparatus defect in reference 23. According to the French data, the observed magnetic splitting is described by a magnetic moment for the excited level equal to zero. This conclusion was obtained on the assumption that there is no quadrupole splitting, and must be reexamined.

Thus the measurements carried out up to the present are not in agreement with one another to a sufficient extent, and do not permit us to draw any definite conclusions concerning the value of  $\mu_1$ .

According to the shell model, the odd neutron in  $\operatorname{Sn}^{119*}$  should be in a  $d_{3/2}$  state. The Schmidt limit in this case is +1.1 n.m.

The measurements should be continued, and to simplify the interpretation it would be desirable to use as source and filter alloys of tin having a lattice with cubic symmetry.

# EXPERIMENTS WITH Zn<sup>67</sup>. FURTHER POSSI-BILITIES

 $Zn^{67}$  has an excited level with an energy of 92 kev and a natural width  $\Gamma = 4.8 \times 10^{-11}$  ev. The relative width of the  $\gamma$  transition of  $Zn^{67}$  ( $\Gamma/E = 5.3 \times 10^{-16}$ ) is three orders of magnitude less than the width of the narrowest line which has been studied up to now, the line in  $Fe^{57}$ . Because of this the observation of the Mössbauer effect in  $Zn^{67}$  is of great interest. At the same time the problem is a difficult one. First of all, because of the relatively high transition energy and the low mass of the radiating nucleus, the intensity of the Mössbauer line should be small in the case of  $Zn^{67}$ . Secondly, the unusual narrowness of the line means that there is great danger of masking of the effect because of broadenings and shifts caused by vibrations, non-uniformities in the material, differences in the structure of the source and the filter. It is sufficient to say that a relative velocity of source and filter equal to 0.15 microns/sec shifts the line by an amount equal to the width of the level.

An attempt to detect resonance absorption in Zn<sup>67</sup> was undertaken by a group<sup>11</sup> of physicists at the Joint Institute for Nuclear Studies and the Physics Institute of the Academy of Sciences. In view of the dangers pointed out above, the experiment was set up as follows: The surface of a sample of metallic zinc enriched to 33% Zn<sup>67</sup> was irradiated with 6.7 Mev protons, and the activity Ga<sup>67</sup> was produced in it via a (p,n) reaction. Ga<sup>67</sup> with a half-life of 78 hours is converted to  $Zn^{67}$ , which in a considerable fraction of the decays is formed in its excited state. The sample was placed in a helium cryostat between pole tips producing an inhomogeneous magnetic field with a maximum intensity of the field in the region of the source equal to 1500 oe (Fig. 5). The resonance absorption effect was detected from the change in intensity of the 93-kev radiation passing through 6 mm of Zn, when the magnetic field is turned on to destroy the resonance absorption. Measurements with and without field were done automatically every 20 - 40 sec. The change in the amplification of the photomultiplier when the field was turned on did not exceed  $10^{-5}$ , as shown by special supplementary measurements.



FIG. 5. Diagram of experimental arrangement<sup>11</sup> for observing resonance absorption in Zn<sup>57</sup>. 1 - sample of enriched Zn; 2 - sample of natural Zn; 3 - face of enriched sample irradiated at the cyclotron; 4 - screen at T = 80° K with pole tips made of Armco iron; 5 - magnetic screen of iron; 6 - pole of the electromagnet; 7 - lead collimators. Samples 1 and 2 were soldered to the bottom of the vessel containing liquid helium. Photomultipliers were surrounded by a screen of permalloy 5 mm thick.

As a result of several series of measurements which were in agreement with one another, it was found that at the temperature of liquid helium switching on the field reduces the counting rate by an amount  $\delta = (2.58 \pm 0.84) \times 10^{-2} \%$ . Control experiments at liquid helium temperature [ filtration of the radiation from Zn<sup>67</sup> with energies 180 and 270 kev, filtration of the radiation at 92 kev through a sample containing one-eighth as much Zn<sup>67</sup> (natural Zn)] and at room temperature (filtering through enriched and normal Zn) – all gave a null result within the limits of accuracy of the measurements. The averaged result of all the control experiments was  $\delta = (0.05 \pm 0.4) 10^{-2} \%$ .

Thus we may assume that the observed effect is actually caused by resonance absorption in  $Zn^{67}$ . Theoretical estimates using the effective Debye temperature of metallic zinc of  $\theta = 213^{\circ}$ K as computed by Kazarnovskii,<sup>6</sup> including the quadrupole splitting in the non-cubic lattice of Zn, give for the conditions of the experiment  $\delta = (\sim 9) \times 10^{-2}$ %, which is only 2-3 times greater than the observed value. The experiments with Zn<sup>67</sup> are continuing.

In the experiments of Pound and Rebka,<sup>24</sup> which were done with an accuracy of 0.1% with natural Zn, the effect of resonance absorption, as expected, did not appear.

A significantly greater value of the resonance effect was obtained by Craig et al.,<sup>25</sup> using for the material of the source and filter zinc oxide, which has a higher Debye temperature ( $\theta \simeq 300^{\circ}$ K). The results of their work<sup>25</sup> are shown in Fig. 11 of Mössbauer's paper (cf. page 869 of the present issue).

An extremely interesting question is whether it is possible to use still narrower resonance lines. At present no low-lying excited states of stable isotopes are known with a lifetime in the interval from  $10^{-5}$  to tens of seconds. In this connection, Tzara in France<sup>26</sup> and Burgov and Davydov<sup>27</sup> in the U.S.S.R. considered the possibility of observing the Mössbauer effect on the isotope Ag<sup>107</sup>, which has an excited level with an energy of 88 kev and a half-life of 40 sec. The broadening of the line as a result of interaction of the magnetic moment of the excited state with the magnetic moments of neighboring nuclei and as a result of other influences leads to a reduction of the resonance cross section by several orders of magnitude; but in the opinion of the authors cited the resonance effect should still be observable.

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

<u>Translator's Comment.</u> A set of four papers\* in Soviet Physics-Uspekhi provides the first complete survey of theoretical and experimental work on the recoilless absorption and emission of gamma rays (the Mössbauer effect).

The first paper begins with an excellent summary of Lamb's theory of the capture of neutrons in a crystal lattice, and then extends the theory to resonance scattering of gamma rays by crystals. Very complete discussions of both the theory and the experiments are given for the temperature effects, hyperfine splitting and polarization of gamma rays, and the Harwell and Harvard tests of general <sup>2</sup> F. L. Shapiro, Элементарная теория эффекта Mёссбауэра (Elementary Theory of the Mössbauer Effect), Press of Physics Institute, Academy of Sciences, Moscow 1960.

<sup>3</sup>I. M. Frank, J. Phys. U.S.S.R. 7, 49 (1943).

<sup>4</sup>R. W. James, Optical Principles of the Diffraction of X-rays, G. Bell and Sons, London, 1948.

<sup>5</sup> M. Blackman, Handbuch der Physik VII/1, 325 (1955).
 <sup>6</sup> M. V. Kazarnovskiĭ, JETP 38, 1652 (1960), Soviet

Phys. JETP 11, 1191 (1960).

<sup>7</sup>R. V. Pound and G. A. Rebka, Phys. Rev. Letters 4, 274 (1960).

<sup>8</sup>B. D. Josephson, Phys. Rev. Letters 4, 341 (1960).

<sup>9</sup>A. Dzyub and A. F. Lubchenko, JETP (in press).

<sup>10</sup>A. C. Melissinos and S. P. Davis, Phys. Rev. **115**, 130 (1959).

<sup>11</sup>S. I. Aksenov et al., JETP 40, 88 (1961).

<sup>12</sup>O. C. Kistner and A. W. Sunyar, Phys. Rev. Letters 4, 412 (1960); I. Solomon, Comptes rendus 250, 3828 (1960).

<sup>13</sup> M. I. Podgoretskii and I. D. Roizen, JETP 39,

1473 (1960), Soviet Phys. JETP 12, 1023 (1961).

<sup>14</sup> M. I. Podgoretskiĭ and A. V. Stepanov, JETP, in press.

<sup>15</sup> H. J. Lipkin, Annals of Physics 9, 332 (1960).

<sup>16</sup> V. A. Lyubimov and A. I. Alikhanov, JETP **38**, 1912 (1960), Soviet Phys. JETP **12**, 1375 (1961).

<sup>17</sup> V. A. Lyubimov and A. I. Alikhanov, Reports of the Second All-Union Conference on Nuclear Reactions at Low and Medium Energies, 1960 (in press).

<sup>18</sup> I. Ya. Barit et al., ibid.

<sup>19</sup> N. N. Delyagin et al., JETP **39**, 220 (1960), Soviet Phys. JETP **12**, 159 (1961).

<sup>20</sup> N. N. Delyagin et al., JETP **39**, 894 (1960), Soviet Phys. JETP **12**, 619 (1961).

<sup>21</sup>N. F. Ramsey, in Experimental Nuclear Physics, ed. E. Segre, J. Wiley and Sons, 1953.

<sup>22</sup> C. H. Townes, Handbuch der Physik 38/1, 377 (1958).
<sup>23</sup> J. L. Picou et al., preprint (1960).

<sup>24</sup> R. V. Pound and G. A. Rebka, Phys. Rev. Letters 4, 397 (1960).

 $^{25}$  Craig, Nagle, and Cochran, Phys. Rev. Letters 4, 561 (1960); Nagle, Craig, and Keller, Nature 186, 707 (1960).

<sup>26</sup> C. Tzara, Comptes rendus **250**, 1466 (1960).

<sup>27</sup> N. A. Burgov and A. V. Davydov, Report to the Second All-Union Conference on Nuclear Reactions at Low and Medium Energies, July, 1960.

Translated by M. Hamermesh

relativity. There is a short discussion of further possible applications of the Mössbauer effect. A bibliography of 95 references provides a good key to the original literature.

The papers by Mössbauer, Pound and Shapiro were originally presented at a conference in Russia in July, 1960. Shapiro's paper begins with a very simple classical discussion of the effect and gives some interesting comments on theoretical problems which are still unsolved. His paper provides a summary of all the Russian work in this field and was revised to cover papers some of which are just appearing in the February, 1961, JETP.