RESONANCE EXCITATION OF AN ISOMER STATE IN Ag^{107} *WITH A MEAN LIFETIME OF 63 SECONDS*

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Resonance excitation of the 93.5 keV isomer state in Ag^{107} with a mean lifetime of 63 sec is realized by means of the Mössbauer effect. Gamma activity induced in a sample of metallic silver as a result of resonance absorption of γ rays from a strong source is observed. The activation was carried out at liquid-helium temperature and a low level of acoustic noise. For sources and absorbers heated in vacuum the measured resonance scattering cross section per atom of a natural isotopic mixture is $(0.7 \pm 0.20) \times 10^{-30}$ cm^2.

SOME three years ago we proposed to use the Mössbauer effect for resonant excitation of long-lived isomer states of nuclei [1]. The possibility of such an excitation was considered almost simultaneously and independently in France [2], although there the estimate for the cross section of this process was patently overestimated and no account was taken of several factors that greatly influence the magnitude of the effect. The realization of such an experiment is quite attractive in connection with the exceedingly small level widths, which have lifetimes of the order of several dozen seconds. By measuring under low background conditions the γ activity of an irradiated specimen located away from the primary γ-ray source, it is possible to note the absorption of as low a fraction of the total γ-ray flux incident on the specimen as 10^{-10}—10^{-11}, whereas in ordinary experiments on the transmission, the absorption of 0.1 per cent of the primary intensity is already difficult to observe.

We have made the first successful experiments on resonant absorption of the isomer state of Ag^{107} with energy 93.5 keV and average lifetime 63 sec, corresponding to a natural level width of 10^{-17} eV.

Before we proceed to describe the experimental set-up and the results, let us consider several factors which make the performance of such an experiment difficult.

1. The interaction between the magnetic moments of the silver nuclei greatly broadens the line. The order of magnitude of the resultant effective line width is determined by the energy of this interaction: $\Delta E = 2\mu_1\mu_2/d^3 \cong 10^{-12}$ eV, where μ_1 —magnetic moment of the ground state of silver (0.1 nuclear magneton), μ_2 —magnetic moment of the excited state, assumed equal to two nuclear magnetons, and d —average distance between neighboring atoms. Because of such a broadening, the effective cross section of resonant excitation decreases by approximately 10^5 times.

2. The presence of paramagnetic impurities can also cause additional broadening of the emission and absorption lines. However, when the impurity concentration is smaller than 10^{-5} this broadening does not exceed the broadening due to the interaction between the magnetic moments of the silver nuclei.

3. Acoustic oscillations in a solid can disturb the conditions of resonant absorption, owing to the relative motion of the nuclei of the source and of the absorber. It is necessary to raise the average difference in the velocities of the source and absorber particles to a level $\Delta v \leq 10^{-7}$ cm/sec.

4. The interaction energy between the magnetic moments of the nuclei and the free electrons in metallic silver, estimated from the hyperfine structure of the spectral lines, amounts to $\sim 10^{-5}$ eV. However, owing to the small number of electrons which are effectively acting at low temperature (4—20°K), this interaction leads to a broadening of the γ line only to a value $\sim 10^{-14}$ eV, which is appreciably smaller than the broadening due to the interaction between the nuclear magnetic moments.

Both the source and the absorber were made of one ingot of metallic silver 99.999 percent pure in the form of plates measuring $0.5 \times 20 \times 30$ mm^3. The sources were produced by irradiating these silver plates with protons of energy 17 MeV in the cyclotron of the I. V. Kurchatov Institute of Atomic Energy. The irradiation time was 10—12 hours at a current ~ 70 μA. The $Ag^{107}(p,n)Cd^{107}$ reaction yielded Cd^{107} with activity up to 3 Ci, which corresponds to emission of $\sim 5 \times 10^9$ 93.5-keV γ

quanta per second (the conversion coefficient for this transition is 20.3 ± 2.1 [3]). The contribution of the γ rays from the Ag^{109} isotopes is under our conditions very small, owing to the large half life of Cd^{109}.

Figure 1 shows the experimental set-up. Source 1, wrapped in a double layer of aluminum foil, and absorber 2, in a single-layer wrapper, were placed in a copper box 3 in direct contact with each other.

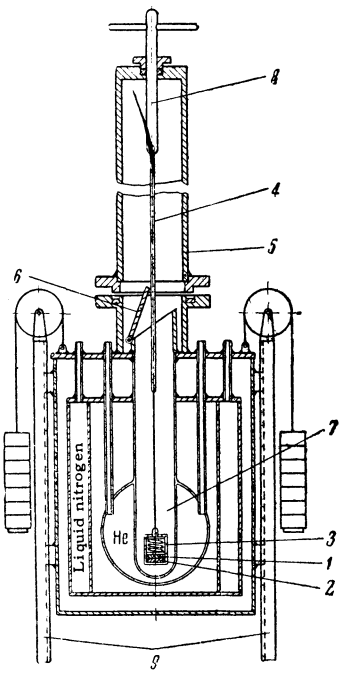

FIG. 1

Box 3, equipped with a spring loaded cover, making it possible to remove the source and absorber rapidly, was suspended on a silon filament ~ 50 cm long and a two-meter rubber cord 4 in a tube 5, the lower part of which was connected through vacuum lock 6 to the inner cavity of a helium cryostat 7. A vacuum-sealed rod 8 in the upper part of tube 5 made it possible to move the box 3 vertically up and down. After dropping the box 3 to the lower part of the cryostat cavity, the air was pumped out of tube 5, which was filled with gaseous helium to a pressure on the order 200—300 mm Hg. Liquid helium was then poured into the cryostat, which was first cooled with liquid nitrogen. The presence of helium in the cryostat, and also the temperature of the box, were determined with carbon resistance thermometers manufactured at the Institute of Physics Problems of the USSR Academy of Sciences [4]. The temperature gradient at the box location in the cryostat cavity did not exceed, according to the measurements, 0.002°K per millimeter, which under our conditions would not lead to any noticeable temperature shift of the lines. After the box was cooled to 4.2°K, the entire set-up was maintained for 3 minutes at the theoretically permissible level of acoustic noise, monitored with the aid of a calibrated acoustic probe which measured the acoustic pressure outside the cryostat. The same probe was used to determine first the ratio between the acoustic pressures inside and outside the cryostat cavity.

The box was then raised rapidly, the vacuum lock 6 closed, and the cryostat, separated from tube 5, was lowered along guides 9. The box 3 was extracted from the tube, opened, and the irradiated specimen placed on a scintillation counter with thin (3 mm) NaI(Tl) crystal in a lead "housing." The source was placed at the same time behind a lead shield. The entire procedure from the end of the irradiation of the plate to the start of the count took 25—30 seconds.

The pulses from the scintillation counter were fed to a single-channel AADO-1 analyzer, tuned to the photopeak of the 93.5-keV line. The width of the channel was 5 V, and the maximum count should have corresponded to a pulse amplitude of approximately 20 V. The counting was carried out continuously for 20 minutes and the readings of the scaler instrument were recorded every minute. The number of counts starting with the fifth minute was used to determine the average background, which amounted to from 5 to 10 counts per minute in different series of measurements. Figure 2 shows the results of 18 series of measurements, obtained by adding 18 count numbers for each succeeding count interval of 1 minute. The shaded strip indicates the average background and its error, calculated using all the points starting with the fifth. It is clearly seen that during the first

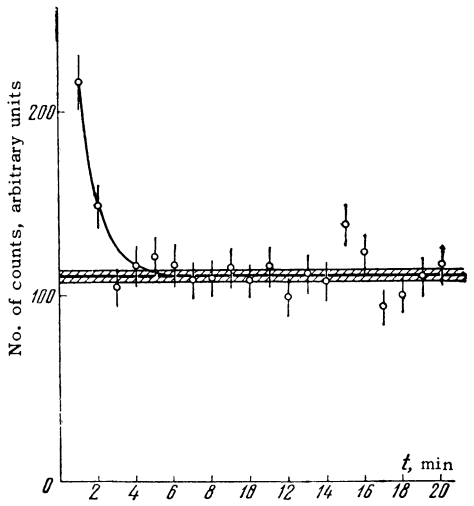

FIG. 2

minute the counting rate greatly exceeds the average background and the decrease in activity occurs in accordance with the average lifetime of the decaying nuclei, equal to 63 seconds (the exponential curve shown in the plot corresponds precisely to this lifetime). It must be noted that annealing the source and the absorber in vacuum for 1.5—2.0 hours at 500—560°C leads to an increase in the resonant absorption by a factor of several times. Apparently the main influence of the annealing consists in restoring the metal crystal structure damaged by proton bombardment, and removal of the hydrogen that has accumulated in the silver during the bombardment process. Annealing of the absorber was aimed at attaining some degree of identity between the two silver plates (the absorber, like the source, was produced by rolling and was expected to contain work-hardened regions).

The cross section for resonant absorption of 93.5 keV γ quanta by silver nuclei can be estimated by calculating the probability of emission (f) and of absorption (f′) of γ rays without recoil. In this case these probabilities should be the same by virtue of the identity of the crystal lattices of the source and absorber (disturbances to the crystal structure due to proton bombardment are disregarded in these calculations).

If the Debye temperature of silver is assumed to be 215°K[5], then the usual calculation in the Debye approximation yields $f = f' = 0.029$ under the condition that the source in the absorber are 4.2°K. At these values of f and f′, the effective resonant scattering cross section per atom of the natural isotope mixture is 0.56 b.

To determine the experimental activation cross section we carried out, in addition to the experiments described above, also measurements of the source activity. The activity was measured directly after the end of the activation experiments. The measurement was made with the same counter and at the same setting of the pulse-height analyzer. The source was secured at a distance of approximately 3 meters from the counter and the number of counts per unit time in a given pulse-height analyzer channel was measured. The absorber could be regarded as thin relative to resonant absorption. Under these conditions, the experimental cross section of resonant scattering was calculated from the following formula:

$$\sigma_{\text{res. scat}} = N_2(t_3)\frac{\exp(\lambda_{Cd}t_1)\exp(\lambda_{Ag}t_2)}{1-\exp(-\lambda_{Ag}t_{\text{ir}})}\left[\int_0^{t_3}\exp(-\lambda_{Ag}t)\,dt\right]^{-1}$$
$$\times\left[n\frac{\pi r^2}{S}\int_0^{d_2}dx'\int_0^{\pi/2}\sin\theta' d\theta'\exp\left(-\frac{x'}{l_\gamma\cos\theta'}\right)\right.$$
$$\left.\times\int_0^{\pi/2}\exp\left(-\frac{x'}{l_\gamma\cos\theta}\right)\frac{\sin\theta}{\cos\theta}N_1(\theta)\,d\theta\right]^{-1}. \quad (1)$$

Here $N_2(t_3)$ is the experimentally determined number of counts during the time from 0 to t_3; t_1 is the time elapsed from the midpoint of the activation experiment to the start of the measurement of the source activity; t_2 —time elapsed from the end of the absorber activation to the start of the measurement of its activity; λ_{Ag}, λ_{Cd} —decay constants for silver and cadmium; t_{ir} —time during which the absorber is activated; n —number of silver atoms per unit volume; S —area of the registering crystal; r —distance from the crystal to the source in the experiment during which the source activity is measured; d_2 —absorber thickness; l_γ —mean free path of 93.5-keV γ quanta in silver; $N_1(\theta)$ is connected with the experimentally determined number of counts per unit time from the source $N_1(0)$ by the relation

$$N_1(\theta) = N_1(0)\left\{\int_0^{d_1}\exp\left(-\frac{x}{l_\gamma\cos\theta}\right)\right.$$
$$\left.\times\sigma_a[E(x)]\,dx\right\}\Big/\left\{\int_0^{d_1}\exp\left(-\frac{x}{l_\gamma}\right)\sigma_a[E(x)]\,dx\right\}. \quad (2)$$

In formula (2) d_1 is the source thickness and $\sigma_a[E(x)]$ the cross section of the $Ag^{107}(p,n)Cd^{107}$ reaction as a function of the depth of proton penetration.

Since the relaxation time of the nucleus spins in the lattice is not known, we have assumed that there is no correlation between the incoming and resonance-scattered γ quantum.

The measured cross section for the resonance scattering of γ rays by Ag^{107} nuclei, for a source and absorber annealed in vacuum, turn out to be $(0.74 \pm 0.20) \times 10^{-30}$ cm^2, which is approximately 7.5×10^5 times smaller than the calculated quantity. The indicated error is statistical. It can be assumed thus that the effective line width is 7.5×10^5 times the natural width, but final conclusions can be drawn only after a direct measurement of the line width.

In addition to the main measurements, several control experiments were performed. In particular, to check whether the observed effect is a result of transport of Ag^{107} recoil nuclei from the surface layer of the silver to the specimen by the vortical streams of gas produced during the dismantling of the cryostat, measurements were made with a copper plate as an absorber. No activation of this plate was noted.

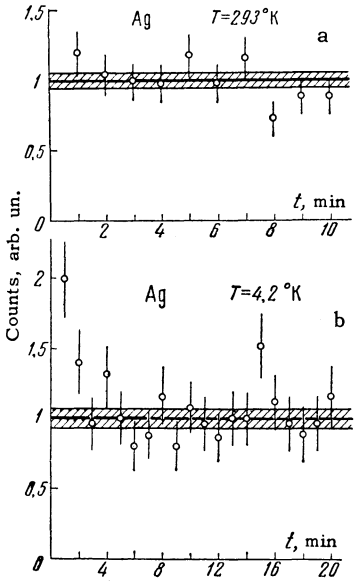

FIG. 3

Figure 3a shows the result of another control experiment. In this case an attempt was made to detect resonant excitation of silver nuclei at room temperature. The absence of excitation at room temperature and its presence at liquid-helium temperature indicates that we actually deal with the Mössbauer effect. Figure 3b shows for comparison the results of excitation of silver nuclei at helium temperature, obtained with the same absorber and source as the data shown in Fig. 3a. These measurements were carried out in succession.

Thus, it can be stated that resonant absorption of a nuclear level with a natural width much smaller than the width of the narrowest level excited previously (Zn^{67}, $\Delta E/E = 5.4 \times 10^{-16}$), has been realized. We are presently undertaking a direct measurement of the effective width of the resonance line.

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[1] A. V. Davydov and N. A. Burgov, Possibility of Excitation of Long-lived Isomer States by Means of the Mössbauer Effect, Report, Institute of Theoretical and Experimental Physics, February, 1960.

[2] C. Tsara, Compt. rend. **250**, 1466 (1960).

[3] Brunner, Huber, Joly, and Maeder, Helv. Phys. Acta **26**, 588 (1953).

[4] N. N. Mikhaĭlov and A. Ya. Kaganovskiĭ, PTÉ, No. 3, 194 (1961).

[5] F. Seitz, The Modern Theory of Solids, McGraw Hill, N.Y., 1940.

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