e.g., by means of apertures inserted in the discharge tube, the wall conditions are modified. This also seems to be supported by experiments.[8] Further, the present theory seems to connect the classical diffusion theory[1,5] and the ion wave instability theory[4] in a natural way.

An important consequence of the present discussions is that a reversal of the radial electric field in a plasma by means of a magnetic field seems to be impossible if the boundary condition Eq. (1) has to be satisfied. Finally, for large values of Rp_0, Eq. (4) shows that the sheath is unstable even in the absence of a magnetic field. This may be of some interest in the study of high-pressure glow discharges.[14]

The author wishes to express his sincere thanks to Professor H. Alfvén, Dr. B. Lehnert, Dr. S. Lundquist, and Dr. E. Åström for many valuable discussions.

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ZEEMAN EFFECT IN THE RECOILLESS γ-RAY RESONANCE OF Zn^{67}†

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Recoil-free resonance emission and absorption (Mössbauer effect)[1] of the 93-kev γ ray in Zn^{67} gives rise to the most precise energy definition thus far reported.[2] Despite the numerous difficulties which beset the experimenter searching for the resonance,[3] a small but definite Mössbauer effect has been found.[2] This Letter reports measurements on the influence of the nuclear Zeeman effect and other perturbing factors upon the Mössbauer effect in Zn^{67} embedded in an enriched ZnO absorber lattice.

The relatively high energy of this γ ray makes it necessary to embed the source and the absorber atoms in rigid crystalline lattices, and to perform the experiments at low temperatures. The first of these requirements was met by using ZnO for both the source and the absorber lattices. The second requirement was more than satisfied by using temperatures below the helium lambda transition (2.175°K). In addition to these basic requirements, several experimental difficulties are consequences of the extreme narrowness of the line (4.84×10^{-11} ev). It is accordingly necessary to take into account the effect of various perturbing influences. Here we list the more important of these perturbations, and in the following paragraphs indicate how they enter into the design of the experiment.

One class of shifts arises from the change of nuclear mass upon γ-ray emission or absorption, with a resultant change in the phonon spectrum of the lattice. The change in energy of the emitted γ ray is given by[4]

$$\Delta E = -(E/Mc^2)\langle T\rangle, \qquad (1)$$

where E is the γ-ray energy, M is the mass of the emitting nucleus, and $\langle T\rangle$ is the expectation value for the kinetic energy per atom of the lattice. If any parameter x should differ between source and absorber lattice, the recoil-free peaks will occur at different energies in the emission spectrum and in the absorption spec-

trum. The shift is given by

$$\delta(\Delta E) = \frac{\delta}{\delta x}(\Delta E)\delta x. \quad (2)$$

This expression predicts that a difference in Debye temperature between source and absorber of only about 1.3°K would cause a shift of one linewidth. Thus a change in the average isotopic mass number of 2% would, through the mechanism of the Debye temperature (taking θ = 300°K), result in a shift of about four linewidths. Similarly, differences in chemical constitution[5] or lattice defects may be expected to produce significant shifts. We shall refer to a shift due to difference in isotopic mass between source and absorber as the isotopic mass effect.

In order for the emitted gamma radiation to remain unshifted by the recoil of the emitting nuclei, it is essential that the recoil momentum be absorbed by entire crystallites. For all previously observed resonances the minimum size of the crystallites was quite small. In the present situation this is no longer true. The recoil momentum must be taken up by at least 2×10^9 nuclei, so that the ZnO crystallites must be larger than 0.4 micron. Since commercial ZnO would normally possess average grain sizes smaller than one micron,[6] care must be exercised to ensure a grain size much larger than the above nominal requirement. A sintering process assured that this requirement was met for the source. A sample of the enriched absorber was studied under an oil immersion microscope. No grains smaller than 0.5 micron were observed, and the majority of the grains were in the range of 1 to 2 microns.

Mechanical vibrations of only 10^{-5} cm/sec would produce a Doppler broadening of about one linewidth. Since the vibration level in our building was nearly 5×10^{-4} cm/sec, the helium cryostat was shock-mounted and the pumping lines carefully decoupled. The source and the absorber were clamped rigidly in a single package, which was suspended by threads in the helium bath. The helium bath was pumped below the lambda transition and the nitrogen radiation shield was frozen to prevent vibrations from boiling liquids and also to place source and absorber in an isothermal bath.

In place of the velocity drive usually used in this type of experiment, the resonance was shifted by means of the nuclear Zeeman effect. The magnetic moment of the Zn^{67} nucleus interacting with an applied magnetic field splits the ground state into six components and the excited state into four components. Selection rules permit twelve component gamma transitions, as is shown in Fig. 1(a). Here $I_g = 5/2$, $I_e = 3/2$, and the ground-state gyromagnetic ratio $g = 0.35$.[7] For the excited state no g value has been measured; we adopt arbitrarily the value -1. (The negative sign is predicted by the shell model.) The source is shielded from the magnetic field and hence the emission spectrum is not split. For simplicity, the effect of quadrupole interactions has been omitted. Assuming a shift δE between source and absorber, resonance can occur for six values of magnetic field. Each component has a width which is compounded of the natural linewidth and the widths due to residual mechanical vibrations, quadrupole broadening, etc. Even if this width is the same for all components, the present method of studying the resonances as a function of an external magnetic field will cause the apparent or magnetic width of each resonance peak to be proportional to the field required to establish the resonance. Thus resonances occurring at low values of the applied field will appear narrower than those established at high fields, and the observed spectrum will appear distorted.

The magnetic field was produced by a small

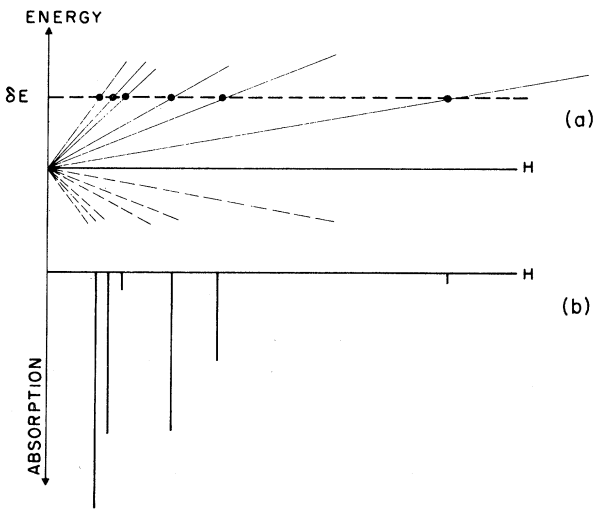

FIG. 1. (a) Zeeman splittings of the gamma ray vs an applied magnetic field. The gyromagnetic ratio of the ground state is 0.35, and a value of -1.0 has been arbitrarily chosen for the excited state. (b) Resonance pattern expected from the splittings in (a) in the presence of an energy shift δE between source and absorber. In actuality, line broadening would be expected to merge closely spaced lines into single peaks.

solenoid surrounding the absorber. Fields of up to 700 gauss could be applied to the absorber, while the source was entirely shielded (at operating temperatures) by superconducting lead foil. Stray fields at the absorber were measured to be less than 2 gauss. Fields from the solenoid were prevented from reaching the photomultiplier by a superconducting ring placed in the cryostat bottom through which the gamma rays emerged. Changes in counting rate due to changing fields at the photomultiplier were found, even without the superconducting shield, to be less than $\pm 0.02\%$.

The absorber used in all experiments was 1.231 g of enriched ZnO (92.4% Zn^{67} enrichment) pressed with 0.036 g of polyethylene glycol binder into a button 1.11 cm in diameter. Sources were prepared in the Los Alamos cyclotron by the reaction $Zn^{66}(d,n)Ga^{67}$ on normal sintered ZnO. Other reaction products were accounted for by a background correction. After bombardment, the sources were annealed for about one hour in air at about 1000°C in order to anneal radiation damage and to assure that the Ga^{67} atoms were correctly placed in the ZnO lattice. Such a procedure is essential, and lower temperatures or shorter times yielded erratic results, or no resonance whatever.

Measurements were made of the transmission of the absorber vs the applied magnetic field. Because of the extremely small change in counting rate and the large number of counts required (typically 10^8 per point), automation of the counting system and extreme system stability were imperative. The 93-kev gamma rays passed through thin windows in the cryostat bottom, and were detected by a NaI(Tl) scintillation crystal. The pulses were amplified and analyzed by a single-channel analyzer followed by a scaler equipped with a digital recorder. In order to eliminate the effect of source decay, all measurements were taken relative to the counting rate when one applied a magnetic field sufficient to destroy the resonance entirely. The magnetic field sequencing and the scaler were controlled by an automatic programmer. Timing was controlled by a thermostatted quartz crystal frequency standard with a stated accuracy of 3 parts in 10^7 per week. The programming was such as to make the measurements insensitive to linear drifts.

Instrument checks were performed by cycling the magnetic field (a) at room temperature, (b) at low temperature using a nonresonant gamma ray, and (c) at low temperature using as an absorber a zinc-containing material (gahnite) in which no resonance exists.[2] In each case the change in counting rate upon application of the magnetic field was found to be zero to within $\pm 0.02\%$.

Figure 2 shows the resonance curve obtained. The points include corrections for background and unresolved nonresonant gamma rays of typically 30%. The central features of the data are the remarkably large total area under the resonance, and the structure and total breadth present. The total area is sufficiently large that, were there no line broadening, one would estimate the resonance absorption to be several percent. Such a value implies a Debye temperature for ZnO of about 300°K, which is consistent with the results obtained from specific heats. The major structural feature of the curve lies in the fact that the maximum resonance does not occur at zero field, but is shifted to about 10 gauss. This result indicates the presence of shifts such as the isotopic mass effect mentioned above. However, the displacement of the maximum (to 10 gauss) is smaller than one would predict from this effect alone unless an unreasonably large value is assumed for the excited state g value. A plausible explanation is that chemical shifts[5] are superimposed upon the isotopic effect. The presence of such shifts is implied by the small resonance found at zero field using an unenriched absorber.[2] Since in that case no isotope effects were present, only chemical shifts or quadrupole broadening can explain the small resonance ob-

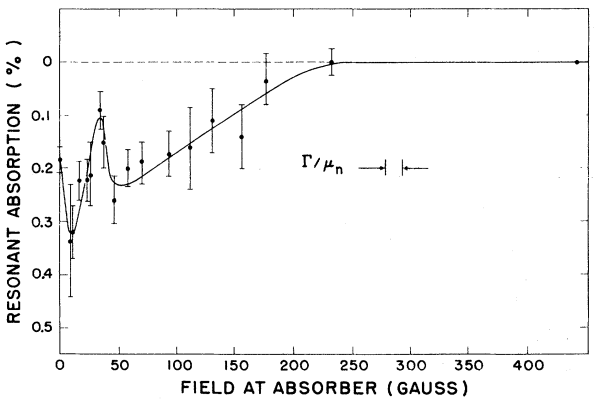

FIG. 2. Experimental resonance absorption pattern for the 93-kev line of Zn^{67} vs the magnetic field applied to an enriched ZnO absorber. The source is in zero magnetic field. Γ/μ_n is the natural level width (in ergs) divided by a nuclear magneton (in ergs per gauss).

served. In the present experiments two peaks are found separated by only about 40 gauss so that it seems unlikely that the broadening is extreme. The tail of the resonance extends to about 400 gauss with no additional structure. This may be related to the characteristic of the magnetic method that even unbroadened lines possess apparent magnetic widths which are proportional to the applied magnetic field.

Although the interpretation is admittedly incomplete, the extreme sharpness of the resonance is apparent. In further study, involving the development of a Doppler shift drive, we hope to measure a number of the energy shifts and level splittings mentioned in previous paragraphs.

We wish to thank S. D. Stoddard and R. E. Cowan for preparation of the ZnO source buttons and for compacting the enriched ZnO absorber. The generous cooperation of the cyclotron group is gratefully acknowledged. W. E. Keller and J. G. Dash each contributed a number of ideas to the experiment.

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OPTICAL AND MICROWAVE-OPTICAL EXPERIMENTS IN RUBY

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Several recent papers[1-4] have reported optical and microwave-optical measurements in ruby (Cr^{+++} in Al_2O_3). We wish to report here some new experiments concerning the fluorescent relaxation processes in this crystal. Reported here also are the first observations of ground-state population changes in ruby due to optical excitation and the detection of optical absorption between two excited states in this crystal.

The predominant processes which ensue in a fluorescent material when it is irradiated at an appropriate wavelength are shown in Fig. 1. W_{13} is the induced transition probability per unit time due to an exciting radiation and the S_{mn} are decay rates which include both radiative and nonradiative processes. In this crystal S_{21} is easily obtained from the decay rate of the fluorescent level (^2E) after an exciting source is turned off. The lifetime for this process is about 5 msec. Varsanyi, Wood, and Schawlow[2] have further demonstrated that this lifetime is almost entirely due to spontaneous emission, i.e., S_{21} is approximately the Einstein A coefficient A_{21}.

An approximate value for the rate S_{32} was obtained in the following way. A crystal of ruby was irradiated with 5600A radiation causing absorption into the lower band ($^4A_2 \rightarrow {}^4F_2$). The sample used was a one-centimeter cube cut from a boule of standard pink ruby supplied by the Linde

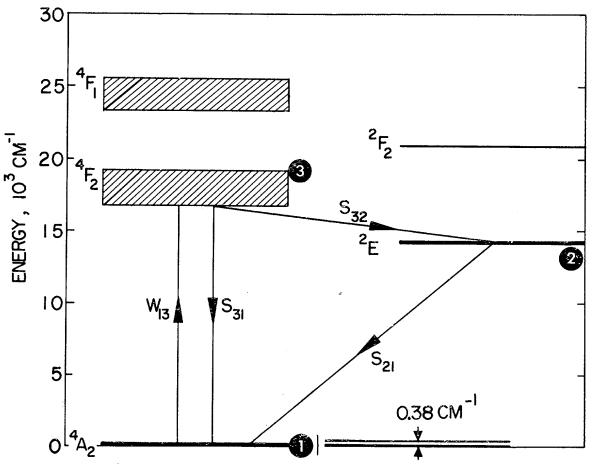

FIG. 1. Pertinent features of the energy level diagram in ruby.