

⁷ Chamberlain, Segrè, Tripp, Wiegand, and Ypsilantis, Phys. Rev. **93**, 1430 (1954), and private communication.

⁸ While these calculations were in progress, Dr. W. Heckrotte sent us preliminary results of calculations of the polarization of 300-Mev nucleons scattered from carbon. Some of these calculations were carried out with the harmonic oscillator potential and gave results similar to those obtained here.

⁹ Fernbach, Serber, and Taylor, Phys. Rev. **75**, 1352 (1949).

¹⁰ T. B. Taylor, Phys. Rev. **92**, 831 (1953).

¹¹ Hofstadter, Fechter, and McIntyre, Phys. Rev. **92**, 978 (1953); L. I. Schiff, Phys. Rev. **92**, 988 (1953).

Statistical Mechanics and the Overhauser Nuclear Polarization Effect*

C. KITTEL

Department of Physics, University of California, Berkeley, California

(Received May 27, 1954)

OVERHAUSER¹ has discovered the remarkable result that under appropriate conditions the population distribution of nuclear spins in a metal among the nuclear magnetic sublevels is determined essentially by the magnitude of the electronic magnetic moment μ_B , rather than by the nuclear moment μ_I . His conditions are that the electron spin resonance of the conduction electrons should be saturated, and that the principal spin-lattice relaxation mechanism of the nuclear spins should be the $\mathbf{I} \cdot \mathbf{s}$ hyperfine coupling with the conduction electrons. The predicted enhancement of nuclear polarization on saturating the electron resonance has been detected experimentally by Carver and Slichter.² Overhauser arrived at his result by a detailed kinetic calculation. The purpose of this note is to establish briefly the connection of the result with the general principles of statistical mechanics and with the second law of thermodynamics.

The canonical ensemble in statistical mechanics describes the distribution of a small sub-system A of a large system $A+B$, as a function of the energy of the remainder B of the system. It is important for our purpose to recall in Gibbsian language the argument which gives the Boltzmann distribution ratio $P^+/P^- = \exp[-2\mu_I H/kT]$ for nuclei of spin $I = \frac{1}{2}$ in thermal equilibrium in a static magnetic field H . The nuclear and electron spin systems will be treated as A , while the reservoir B consists of the crystal lattice and the translational energy of the conduction electrons. We consider what happens when one nuclear spin is reversed. With $\mathbf{I} \cdot \mathbf{s}$ coupling dominant,³ an electron spin must simultaneously turn the other way. The energy change of the combined spin systems is $2(\mu_B - \mu_I)H$ and is balanced by a corresponding change $-2(\mu_B - \mu_I)$ in the translational energy of a conduction electron. We are interested in the effect of a nuclear spin flip alone, and therefore we are to look at the system at a later time when the reversed electron has been restored to its original direction with energy change $-2\mu_B H$ by

means of a relaxation process⁴ not involving the nuclear spin. The net effect of the reversal of a nuclear spin is to change the energy of the reservoir B by $-2(\mu_B - \mu_I)H + 2\mu_B H = 2\mu_I H$. This energy change in the reservoir B changes the volume of phase space accessible to the reservoir, and the later change gives directly the canonical distribution of the sub-system A . It is to be noted that we have no interest in processes in which the restoration of the original electron spin orientation is accompanied by the restoration of the nuclear spin.

When the system is not in thermal equilibrium, but is flooded with rf power to saturate completely the conduction electron spin resonance, the corresponding net energy change in the reservoir on reversing one nuclear spin is now $-2(\mu_B - \mu_I)H$, instead of $2\mu_I H$. The reason is simple: the restoration of the electron spin to its original direction now occurs by the action of the rf field, instead of by a relaxation process. This is the significance of rf saturation. The magnetic energy change $-2\mu_B H$ is provided by the rf field instead of by the reservoir B . The reservoir had its energy changed by $-2(\mu_B - \mu_I)H$ at the beginning when the nuclear and electron spins flipped at the same time, and this is now the only energy change suffered by the reservoir in connection with the total process. The distribution of the sub-system reflects the energy change $-2(\mu_B - \mu_I)H$ of the reservoir, so that the population of the nuclear magnetic sublevels is now given by $P^+/P^- = \exp[2(\mu_B - \mu_I)H/kT]$ under rf saturation of the electron resonance. This demonstrates the enhancement of the nuclear polarization and agrees with the original result of Overhauser.

The discussion may be generalized to treat polarization of a nuclear spin system which suffers spin-lattice relaxation by dipolar or hyperfine interaction with an rf-saturated electron spin system. We let f_B denote the fraction of the spin-lattice relaxation processes of the nuclear spin system which are accompanied by an electron spin change in the opposite sense ($I-S^+$ type interaction), while f_F denotes the fraction accompanied by an electron spin change in the same sense ($I-S^-$ type interaction). Then the nuclear population ratios are

$$P^+/P^- = \exp\{2[(f_B - f_F)\mu_B - \mu_I]/kT\},$$

under saturation conditions, when $S = I = \frac{1}{2}$. As noted also by Bloch, the enhancement effect is not necessarily restricted to metals. It may even be possible to observe the effect in materials with a low concentration of single unpaired electrons. The irradiation of hydrogenous material might introduce suitable centers for proton polarization.

It may be noted that saturation of the spin resonance associated with a zero-field splitting may also enhance polarization. We exhibit the simplest case, taking $f_B = 1$, $S = 1$, and an electronic zero-field splitting ΔE in an axial crystalline electric field. Then $P^+/P^- = \exp[\Delta E/kT]$, provided that the resonance at

$h\nu = \Delta E$ is saturated. Nuclear electric quadrupole splitting of a second nuclear species with $I=1$ could be substituted for the electronic system, if ΔE were large enough to be useful.

I am indebted to Professor F. Bloch for helpful discussions.

* Assisted in part by the National Science Foundation.

¹ A. W. Overhauser, Phys. Rev. **92**, 411 (1953); an elementary kinetic derivation of Overhauser's principal result has been given by F. Bloch, Stanford meeting of the American Physical Society [Phys. Rev. **93**, 944 (1954)].

² T. R. Carver and C. P. Slichter, Phys. Rev. **92**, 212 (1953).

³ W. Heitler and E. Teller, Proc. Roy. Soc. (London) **A155**, 637 (1936); see also J. Korryng, Physica **16**, 601 (1950).

⁴ It is thought at present that the most important process of conduction electron spin relaxation may be by spin reversal during electron collisions with phonons and lattice imperfections, as discussed by R. J. Elliott, Phys. Rev. (to be published).

Nuclear Scattering of Gamma Rays below Meson Threshold*

G. E. PUGH,† D. H. FRISCH, AND R. GOMEZ

Department of Physics and Laboratory for Nuclear Science,
Massachusetts Institute of Technology, Cambridge, Massachusetts

(Received June 1, 1954)

USING an energy-sensitive gamma-ray detector (which will be described soon in an article for the *Review of Scientific Instruments*) we have made preliminary measurements, at 90° and 135° , of the absolute gamma-ray scattering cross section for Be, C, Al, Cu, Sn, Pb, and Bi in the energy range from 35 to 130 Mev. The work was done in the bremsstrahlung beam of the M.I.T. synchrotron using targets about $\frac{1}{4}$ radiation length thick. The maximum beam energy was kept just below meson threshold to prevent confusion with decay gamma rays from the π^0 meson.

A standard coincidence, anticoincidence telescope with a lead converter was used to identify the gamma ray. The telescope was followed by a very large liquid scintillator which integrates the energy loss of the electron pair in its volume and thus estimates the energy of the gamma ray. The pulse height response of the counter to monoenergetic events, ranging from 25 to 150 Mev, was measured using electrons of known energy to simulate gamma rays (note the family of curves in Fig. 1A).

By using the known bremsstrahlung spectrum of the synchrotron, the efficiency of the converter, and the measured response of the counter to any energy event, it is possible to predict the response of the counter for any arbitrary scattering cross section.

In exact analogy with atomic x-ray scattering, the differential cross section was taken as: $d\sigma/d\Omega = \sigma^0 [Z^2 f^2 + (1-f^2)Z]$, where σ^0 is the individual-particle cross section and f is the nuclear analog of the atomic struc-

ture factor given by

$$f = \int_0^\infty \frac{\sin kr}{kr} \frac{4\pi r^2 \rho(r)}{Z} dr.$$

Response curves were computed for a uniform distribution [$\rho(r) = \text{const}$] of protons in nuclei of radii $R = R_0 A^{\frac{1}{3}} \times 10^{-13}$ cm. In Fig. 1 the curves (a) $R_0 = 0.8$, (b) $R_0 = 1.1$, (c) $R_0 = 1.4$ were obtained by using for σ^0 the classical Thompson individual-proton cross section:

$$\sigma_p^0(\text{Thompson}) = \frac{1}{2} (e^2/mc^2)^2 (1 + \cos^2\theta).$$

For elements heavier than aluminum the agreement with this classical Thompson scattering is as good as the statistics of the experiments. The results point to $R_0 = 1.1 \pm 0.2$.

In the lighter elements there is a clear disagreement which corresponds to too large a backward scattering of high-energy photons. Careful analysis of the data on Be and C enables us to deduce an experimental σ^0 which gives a best fit to the data, when R_0 is taken as 1.1. The ratio of this $\sigma_{(n+p)}^0$ "obs" to σ_p^0 Thompson together with a rough estimate of its statistical band of error is plotted in Fig. 2. The statistics are far from conclusive, but the trend of the data suggests that the

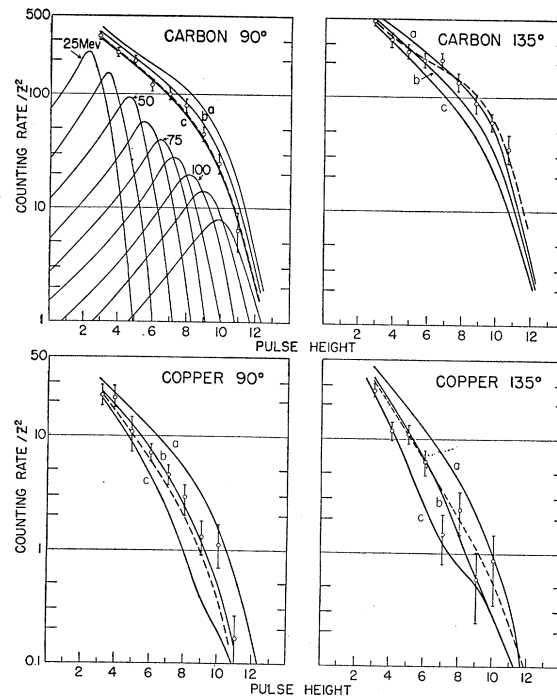


Fig. 1. Typical data for one light and one heavy element. Curves *a*, *b*, and *c* are theoretical response curves of counts vs pulse height computed using Thompson scattering only by a uniform distribution of free, classical protons in a nucleus of radius $R = R_0 A^{\frac{1}{3}} \times 10^{-13}$. (a) $R_0 = 0.8$; (b) $R_0 = 1.1$; and (c) $R_0 = 1.4$. The dotted curves are identical with (b) except that they use the modified Thompson cross section $\sigma_{(n+p)}^0$ "obs" shown in Fig. 2. The theory and experiment are on the same absolute scale; neither ordinate nor abscissa is normalized arbitrarily.