ter of the spin resonance line;  $T_2$  is the spinspin relaxation time. In the steady state

$$\Delta \mathbf{N} = \frac{\Delta \mathbf{N}_0}{1 + (2\pi |\mathbf{G}|^2 \mathbf{e}_{\mathbf{X}\mathbf{Z}}^2 / \mathbf{M}^2) \mathbf{T}_1 \mathbf{T}_2}, \qquad (4)$$

so that saturation effects are important when

$$e_{XZ}^{2} \approx \hbar^{2} / 2\pi T_{1} T_{2} |G|^{2}$$
 (5)

If we set  $T_1 \sim 10^{-5}$  sec,  $T_2 \sim 10^{-6}$  sec,  $|G|^2 \sim 10^{-27}$  erg<sup>2</sup>, we have  $e_{XZ} \sim 3 \times 10^{-9}$ , corresponding to a phonon energy flux of about 0.1  $\mu$ watt/cm<sup>2</sup>. The photon energy flux required for saturation is also ~ 0.1 watt/cm<sup>2</sup>, but in an actual experiment this will be reduced by the Q of the cavity. We note from Eq. (1) that  $T_1 |G|^2$  in Eq. (5) is relatively insensitive to the precise nature of the paramagnetic center. The ultimate sensitivity is determined by the sensitivity to saturation changes; if  $10^{-3}$  of saturation can be detected, one can detect a phonon flux ~  $10^{-4} \mu$ watt/cm<sup>2</sup>.

I am indebted to C. H. Townes, R. Truell, and K. M. Watson for helpful discussions.

<sup>\*</sup>This research was supported in part by the Office of Naval Research, the Signal Corps, the Air Force Office of Scientific Research, and the National Security Agency.

<sup>1</sup>Some of the difficulties in detecting microwave phonons are discussed by C. Kittel, J. Acoust. Soc. Am. 21, 308 (1949).

<sup>2</sup>Private communications from J. K. Galt, J. R. Pellam, and E. M. Purcell.

<sup>3</sup>See, for example, N. Tsuya, Science Repts. Ritu <u>B8</u>, 161 (1957), who deduces from experimental magnetostrictive measurements values of G ~  $10^{-14}$  to  $10^{-13}$  erg for various ferrites.

## SPIN-LATTICE RELAXATION RESONANCES IN SOLIDS\*

 H. S. Gutowsky and D. E. Woessner<sup>†</sup> Noyes Chemical Laboratory, University of Illinois Urbana, Illinois (Received June 2, 1958)

This letter discusses a spin-lattice relaxation mechanism, which has some novel features, and describes preliminary experiments<sup>1</sup> which suggest the importance of the mechanism in certain cases. Possible applications of the mechanism to spin-echo storage memories and to masers are outlined. If the lattice motions producing spin-lattice relaxation are random and thus have a Debye spectral density distribution with a correlation time  $\tau_c$ , the dependence of  $T_1$  upon the nuclear resonance frequency  $\omega$  has the form<sup>2</sup>

$$(1/T_1) \propto C \tau_c / (1 + \omega^2 \tau_c^2).$$
 (1)

Major deviations from Eq. (1) are possible in situations such as the following. Consider a single crystal containing two nuclear species, one with  $I = \frac{1}{2}$ , and the second with  $I > \frac{1}{2}$  and subject to an electric field gradient. The arguments are general but we will assume the first species to be protons and the second,  $C1^{35}$  for which I = 3/2. In a static magnetic field,  $H_0$ , the two proton levels are separated by  $\gamma_1 \not M_0$ . On the other hand, the chlorine nuclei have four levels which arise from the Zeeman splitting<sup>3</sup> of the doubly degenerate pure quadrupole levels,  $E_m^0$ . For an axially symmetric field gradient, the levels are given to first order in  $H_0$  as

$$\mathbf{E}_{\mathbf{m}} = \mathbf{E}_{\mathbf{m}}^{\mathbf{0}} + \mathbf{m} \gamma_2 \mathbf{h} \mathbf{H}_0 \cos \theta \qquad \mathbf{m} \neq \frac{1}{2} ,$$

$$\mathbf{E}_{\mathbf{m}} = \mathbf{E}_{\mathbf{m}}^{\mathbf{0}} + \mathbf{m}\gamma_2 \, \mathbf{M}_{\mathbf{0}} [\, (\mathbf{I} + \frac{1}{2})^2 \sin^2 \theta \qquad (2)$$

$$\begin{array}{c} +\cos^{2}\theta \end{bmatrix}^{1/2} \qquad m=\frac{1}{2} , \\ \text{where} \\ E_{m}^{0} = \frac{1}{4} \left[ e^{2}qQ/I(2I-1) \right] \left[ 3m^{2} - I(I+1) \right] . \end{array} \tag{3}$$

Thus, the separations of the chlorine levels depend on the quadrupole coupling,  $e^2qQ$ , on  $H_0$ , and also on the angle  $\theta$  between  $H_0$  and the symmetry axis of the field gradient.

Because of the different factors governing the energy levels for protons and chlorine nuclei it is possible to find cases where, by adjusting  $\theta$ and H<sub>0</sub>, the proton resonance frequency can be brought into coincidence with a chlorine transition. While this "resonance" condition is satisfied, proton-chlorine spin exchange can occur. Usually, the quadrupolar coupling of nuclear spins with the lattice is much stronger than magnetic coupling. Therefore the spin exchange, which transfers energy between protons and thence to the lattice, can reduce the proton T<sub>1</sub> by as much as several orders of magnitude.

A schematic representation of the two sets of levels and their dependence on  $H_0$  is given in Fig. 1. Two simple types of experiment can be performed to characterize the relaxation mechanism. With a single crystal in a given orientation, the proton  $T_1$  may be observed as a func-



FIG. 1. A schematic representation of the proton and  $C1^{35}$  energy levels, as a function of applied magnetic field, for a single crystal in a given orientation. Spin exchange occurs between the proton levels and the +3/2, -1/2'  $C1^{35}$  levels when  $H_0 = H_C$ , as shown by the solid arrows. The populations of both of these pairs of levels can be inverted at  $H_0 = H_C$  by saturating the  $C1^{35}$ - $3/2 \rightarrow -1/2'$  transition, shown by the dashed lines.

tion of  $H_0$  and thus of  $\omega$ . We should find the slow change in  $T_1$  predicted by Eq. (1) and depending upon  $\theta$  and  $E_m^0$  there will be up to four very sharp deep minima<sup>4</sup> corresponding to the four high-frequency Cl<sup>35</sup> transitions shown in Fig. 1. Or  $H_0$  could be kept constant and  $T_1$  plotted <u>ver-</u> <u>sus</u>  $\theta$ , obtaining minima much as before. The decrease in the proton  $T_1$  will depend upon the distances between protons and Cl<sup>35</sup> nuclei, and also upon angular factors determining the spin exchange probabilities.<sup>5</sup> The limiting value for the proton  $T_1$  will, of course, be approximately the value of  $T_1$  for Cl<sup>35</sup>.

Experiments demonstrating the existence of the above mechanism were performed on solid p-dichlorobenzene and p-bromochlorobenzene at room temperature using rf pulse apparatus.<sup>6</sup> The normal proton  $T_1$  in these samples is  $10^4$ sec while T<sub>1</sub> for Cl<sup>35</sup> is about  $2 \times 10^{-2}$  sec. At a given time only a small fraction of the molecules in the polycrystalline samples used can have orientations such that a chlorine transition coincides with the proton frequency. However, by rotating the sample in H<sub>0</sub> during the proton relaxation experiment, most of the protons can be brought into resonance with chlorines and relax by spin exchange. It was found that rotation of the cylindrical samples at 30 rpm decreased the proton  $T_1$  to values as short as 2.5 sec, depending upon the proton frequency. The observed dependence of  $T_1$  upon frequency is complex because of the polycrystalline samples, the presence of Cl<sup>37</sup> as well as of Cl<sup>35</sup>, and the rotational averaging. It is significant that the rotational effect was not found at proton frequencies below 23.22 Mc/sec. This is the lowest frequency at which a chlorine transition (Cl<sup>37</sup>) can coincide with the proton frequency, as calculated with Eq. (2) using the Cl<sup>37</sup> pure quadrupole resonance frequency of 27.00 Mc/sec.

It appears that this relaxation mechanism may prove useful in two types of application. Anderson et al.<sup>7</sup> showed that spin echoes can be used as a serial-storage memory device. However, the T<sub>1</sub> of the sample determines the storage time as well as erasure, and in their experiments they could only pre-set  $T_1$  over a limited range by selecting the sample. It now appears possible to adjust the T<sub>1</sub> and the storage time over a very wide range simply by varying  $H_0$  or  $\theta$  for a properly chosen single crystal. Erasure can be accomplished at any time by adjusting  $H_0$  or  $\theta$  momentarily to optimize the resonance relaxation. Moreover, the control and adjustment operations can be performed with simple electrical circuits.

The second possible application concerns proposals<sup>8,9</sup> made for three-level nuclear quadrupole rf masers. Unfortunately, such devices are on the borderline of feasibility because the power available is proportional to  $\gamma^2$ , and  $\gamma_Q$  is relatively small for quadrupolar nuclei. However, suppose the operating frequency represents a transition between levels which can undergo spin exchange with proton magnetic levels. Inversion of the populations of the quadrupole levels is transmitted thereby to the proton levels and both sets of levels contribute to the stimulated emission. Thereby the power available may be increased by a factor up to  $(\gamma_{\rm H}/\gamma_{\rm Q})^2$  which is between 20 and 100 depending on the nuclear species involved. This situation is shown schematically in Fig. 1.

<sup>†</sup>Esso Research Company Postdoctoral Fellow in Chemistry.

<sup>1</sup>A report of more detailed studies will be submitted by D. E. Woessner and H. S. Gutowsky to J. Chem. Phys.

<sup>2</sup>Bloembergen, Purcell, and Pound, Phys. Rev. 73, 679 (1948).

<sup>\*</sup>This research was supported in part by the National Science Foundation and by the Office of Naval Research.

<sup>3</sup>C. Dean, Phys. Rev. 96, 1053 (1954).

<sup>4</sup>At the  $T_1$  minima, both nuclear species will contribute to the resonance observed. In principle, different  $T_1$ 's should be resolvable for the two species; however, the proton signal will usually be much stronger than that of the other species.

<sup>5</sup>The spin exchange probabilities will also depend upon the relative signs of  $\gamma_1$ ,  $\gamma_2$ , <u>q</u> and <u>Q</u>.

<sup>6</sup>Buchta, Gutowsky, and Woessner, Rev. Sci. Instr. 29, 44 (1958).

<sup>7</sup>Anderson, Garwin, Hahn, Horton, Tucker, and Walker, J. Appl. Phys. <u>26</u>, 1324 (1955); <u>27</u>, 196 (EV) (1956).

<sup>8</sup>R. Braunstein, Phys. Rev. <u>107</u>, 1195 (1957). <sup>9</sup>J. Itoh, J. Phys. Soc. Japan <u>12</u>, 1053 (1957).

## INSTABILITY, TURBULENCE, AND CONDUCTIVITY IN CURRENT-CARRYING PLASMA

O. Buneman Stanford Electronics Laboratories, Stanford University, Stanford, California (Received June 2, 1958)

In fully ionized plasma, close (large-angle) collisions between particles in appreciable relative motion are rare. However, collective Coulomb interactions (i.e., small-angle collisions) cause instabilities which grow so rapidly that relative motions of ions and electrons, i.e., currents, are continually damped down by con-

$\beta$ u- $\omega_{ m pe}$	-2.0	-1.5	-1.0
Rew	0.05	0.09	0.15
${ m Im}\omega$	0.48	0.54	0.60

All quantities in the table are in units of  $\omega_{pi}^{2/3} \times \omega_{pe}^{1/3}$ . The growth rate,  $\text{Im}\omega = (\omega_{pi}^{2}\omega_{pe}\cos\theta)^{1/3} \times \sin\theta$ . maximizes at  $\theta = \pi/3$ , where  $\beta u = \omega_{pe}$ .

For hydrogen,  $\omega_{pe} = 43 \,\omega_{pi}$  and the shortest e-folding times of fluctuation amplitudes are  $18\omega_{pe}$ . (Multiply by  $A^{1/3}$  for ions of mass number <u>A</u>.) The wavelength is then the distance traveled by the electrons during one electron-plasma period. But appreciable growth occurs over a band of wavelengths 20% longer to 10% shorter (for hydrogen). Exponential rise is accompanied by very little oscillation (small Re $\omega$ ). Energy doubles in just about one electron plasma period. The growth rate is independent of u, but

8

version of directed energy into (random) fluctuation energy.

The growth mechanism is the familiar <sup>1-4</sup> two-stream amplification. For initially stationary ions of uniform charge density  $\rho$  [ plasma frequency  $\omega_{\text{pi}} = (e \rho / M \epsilon /_0)^{1/2}$ ] traversed by electrons [charge density  $-\rho$ , plasma frequency  $\omega_{\text{pe}} = (e \rho / m \epsilon_0)^{1/2}$ ] with velocity <u>u</u>, the (real) wavelength  $2\pi/\beta$  of fluctuations and their (possibly complex) frequency  $\omega$  obey the dispersion formula

$$\omega_{\rm pi}^2/\omega^2 + \omega_{\rm pe}/(\beta u - \omega^2) = 1.$$

[A fluctuation Coulomb field  $\vec{E}$  creates ion velocities e  $\vec{E}/i\omega M$  and, by virtue of continuity, ion charge density fluctuations  $-(\rho/i\omega)\vec{\nabla}\cdot e\vec{E}/i\omega M = \epsilon_0 \vec{\nabla}\cdot \vec{E} \omega_{pi}^2/\omega^2$ . Similarly the electrons contribute charge density fluctuations  $\epsilon_0 \vec{\nabla}\cdot \vec{E} \omega_{pe}^2/(\beta u - \omega)^2$ , encountering the fluctuations at the Doppler-shifted frequency  $\beta u - \omega$ . Poisson's equation furnishes the dispersion law.]  $2\pi/\beta$  is measured along the electron flow; transverse wavelengths are irrelevant.

Exploring the likely range  $\omega_{pi} \ll |\omega| \ll \omega_{pe}$ for possible complex  $\omega = |\omega| \exp(i\theta)$ , one equates first the imaginary parts in the approximation

$$\beta u/\omega_{pe} - \omega/\omega_{pe} = (1 - \omega_{pi}^2/\omega^2)^{-1/2} \approx 1 + \omega_{pi}^2/2\omega^2$$

giving  $|\omega|^3 = \omega_{pi}^2 \omega_{pe} \cos \theta$ . Then, from the range  $0 < \theta < \pi/2$  and the real part one deduces the following table.

-0.5	0	0.5	1.0	1.5
0.25	0.40	0.58	0.78	1.00
0.66	0.69	0.66	0.51	0

our arguments imply that  $\underline{u}$  exceeds random electron velocities.

If a field is briefly applied to a plasma in thermal equilibrium, accelerating electrons to energy <u>eV</u> within an interval shorter than a plasma period, initial thermal energy <u>eV<sub>0</sub></u> of plasma oscillation within the relevant wavelength band will amplify sufficiently to destroy the directed electron motion in about  $\log_2(V/V_0)$ electron-plasma periods (probably 10-20 for typical cases).

When a quasi-steady current is maintained by a lasting applied field  $\overline{E}$ , fluctuations will never be allowed to drop appreciably below the level