

TABLE I. The total interaction cross section of 1.0-Bev negative pions with H and (D-H). Measurements are listed for absorbers of various hydrogen or deuterium content and for several rms values θ_{rms} of the half-angle subtended by counter No. 5 at the absorber.

θ_{rms}	Absorber g cm ⁻² of H	$\sigma(\pi^-,p)$ mb	Absorber g cm ⁻² of D	$\sigma(\pi^-,d-p)$ mb
7.2°	8.27	39 ±3	13.34	17 ±3
5.4°	3.90	42 ±4.5
3.2°	12.17	49 ±3
2.7°	13.34	21.2 ±2.6
2.5°	3.90	47.5 ±5
2.2°	12.17	46 ±3.5

in a previous letter¹ was placed in the 1.1-Bev negative pion beam which, like the 1.55-Bev beam, is deflected by the Cosmotron magnet. The method for measuring the cross sections was the same as that described before.

In this beam the muon contamination was larger than at 1.55 Bev. It was determined by two arrangements; both make use of the strong-pion and weak-muon nuclear interaction so that, after passing through thick absorbers, the beam contains most of the initial muons and comparatively few pions. The counting rate beyond the absorber has to be corrected for the pions which remain and for the loss of muons by multiple Coulomb scattering in the absorber. In the first arrangement, an absorption curve in Al was obtained up to 307 g cm⁻², by placing the Al between counters 4 and 5. The correction for the loss of muons by scattering was calculated in this case to be 17 percent. In the second arrangement, counters 4 and 5 were arranged as for the CH₂-C measurements with the 73 g cm⁻² of C in place. In addition, 372 g cm⁻² of Fe was placed behind counter 5. A single large counter was placed behind the Fe and was used in several successive positions to integrate the remaining beam over an effective diameter of 14 in. In this arrangement, the loss of muons by scattering was negligible, but a somewhat greater uncertainty than that of the first method was involved in placing safe limits on the remaining pions. The result of the first method is $I_\mu/I_0 = 0.110 \pm 0.025$ while the second gives $I_\mu/I_0 = 0.117 \pm 0.012$.

Two thicknesses of absorber have been used in the CH₂-C measurements (12.17 and 3.9 g cm⁻² of H). The thick absorber introduces less statistical uncertainty and an effectively larger muon correction than the thin absorber. The muon contamination produces a correction in $\sigma(\pi^-,p)$ of 8 mb for the thin absorber and 12 mb for the thick. The results are in agreement, and quoted errors include the uncertainty in evaluating the muon contamination. The electron contamination has been measured as at 1.5 Bev, and found to be less than 1 percent.

Several geometries have been used to check that the cross-section values obtained are equal to the total cross section; that is, that the fraction of events in which secondaries enter our last counter is, indeed, very small with the geometry most extensively used. Taking account of the differences in solid angle, the data reported in Table I show that $\sigma(\pi^-,p) = 48 \pm 4$ mb at 1.0 Bev, which is distinctly larger than the value of 27.5 ± 6 mb obtained by Lindenbaum and Yuan² at 450 Mev and, with reasonable certainty, exceeds our value of 34 ± 3.5 at 1.5 Bev.¹ The value of Shapiro, Leavitt, and Chen³ of $\sigma(\pi^-,p) = 47 \pm 5$ mb at 850 Mev fits quite smoothly on a curve showing a broad maximum at roughly 1 Bev.

Another interesting feature shown by our data is the ratio between $\sigma(\pi^-,p)$ and $\sigma(\pi^-,d-p)$. $\sigma(\pi^-,d-p) = 21 \pm 3$ mb and should be equal to $\sigma(\pi^+,p)$ if the principle of charge symmetry and the additivity of cross sections in deuterium were rigorously correct. While actually some difference can be expected, principally because of the second assumption, the qualitative fact that $\sigma(\pi^-,p)$ is considerably larger than $\sigma(\pi^+,p)$ at 1 Bev can hardly be doubted. Preliminary measurements with positive pions at 800 Mev support our present conclusion.

From $\sigma(\pi^-,p)$ and $\sigma(\pi^+,p)$ one can derive the value of the cross section for the pure state having total isotopic spin of one-half:

$\sigma(\pi^-,p)_{I=1/2} = \frac{3}{2}\sigma(\pi^-,p) - \frac{1}{2}\sigma(\pi^+,p)$. It is 62 ± 7 mb at 1 Bev and about one-half of this value at both 0.45 Bev and 1.5 Bev. This state seems, therefore, to have a very marked maximum at about 1 Bev.

We acknowledge many stimulating conversations which we have had with R. Serber, C. N. Yang, W. Rarita, and H. S. Snyder. C. F. Woolley, Jr., has most valuably assisted us throughout these experiments.

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† On leave from The Johns Hopkins University, Baltimore, Maryland.

¹ Cool, Madansky, and Piccioni, Phys. Rev. **93**, 249 (1954); Bull. Am. Phys. Soc. **28**, No. 6, 14 (1953).

² S. Lindenbaum and L. Yuan, Phys. Rev. **92**, 1578 (1953).

³ Shapiro, Leavitt, and Chen, Phys. Rev. **92**, 1073 (1953).

Transient Nuclear Induction Signals Associated with Pure Quadrupole Interactions*

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WE have observed transient "pure quadrupole" induction signals corresponding to the "Bloch decays" and echoes found in pulsed nuclear magnetic resonance experiments.¹ The induction signals arise from oscillating components of magnetization established along the axis of the applied rf magnetic field H_1 . A quantum-mechanical calculation predicted that these transient components should occur even though, at equilibrium, the electric interaction $Q \cdot \nabla E$ produces no macroscopic magnetization. The presence of the induction decays had been suggested by Dean² to explain the anomalous signal to noise behavior of his quenched oscillator in an earlier investigation of pure quadrupole spectra. We have found the predicted induction signals in NaClO₃ single crystals and powders, following the application of rf pulses at the Cl³⁵ pure quadrupole resonance frequency (29.920 Mc/sec at room temperature). We observe the effects of nuclear Zeeman splittings upon the induction signals by orienting the single crystals within a small magnetic field.

To develop a theoretical expression for the induction signals, we consider a Hamiltonian of the form $\mathcal{H} = Q \cdot \nabla E + \mu \cdot (H_0 + H_1 \cos \omega t)$, where H_0 and H_1 are the amplitudes respectively of the applied dc and rf magnetic fields. We expand the wave function of the Cl nuclei (spin $\frac{3}{2}$) in terms of the eigenfunctions of the electric quadrupole term, $\psi = \sum C_m \exp(-iE_m t/\hbar) \psi_m$. The induction signals observed arise from the components along H_1 of the bulk magnetization $M_{x(y)}$, which is created transverse to the symmetry axis of $\nabla E \cdot M_{x(y)} \propto \bar{I}_{x(y)} = (\psi^* I_{x(y)} \psi)$, where the $I_{x(y)}$ are the transverse components of the nuclear angular momentum operator.

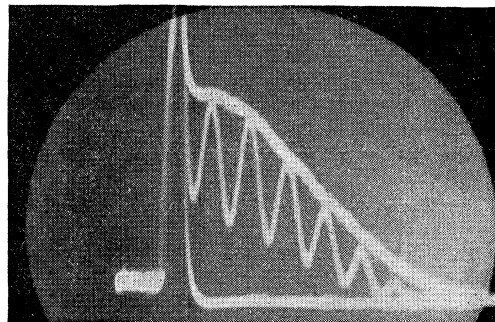


FIG. 1. Induction decay in a single crystal of NaClO₃. The sweep is 800 μ sec long. The beat structure appears in the presence of a Zeeman field of 12 gauss applied, parallel to H_1 , along (0,0,1).

For spin $\frac{3}{2}$, $\bar{I}_{x(y)} \propto (C_{\frac{1}{2}}^* C_{\frac{3}{2}}(\pm) C_{-\frac{3}{2}}^* C_{-\frac{1}{2}}) \exp(i\omega t) + \text{complex conjugate}$, where $\omega = (E_{\frac{1}{2}} - E_{\frac{3}{2}})/\hbar = (E_{-\frac{1}{2}} - E_{-\frac{3}{2}})/\hbar$.

We solve the time-dependent Schroedinger equation in two intervals: during the pulse, in the presence of the rf field H_1 , and following the pulse, where H_1 is zero. The effect of H_1 is then to induce only the transitions $\frac{3}{2} \rightleftharpoons \frac{1}{2}$ and $-\frac{3}{2} \rightleftharpoons -\frac{1}{2}$. Considering the Boltzmann surplus and assuming the system to be initially in the $\pm \frac{3}{2}$ state, the result (for $H_0=0$) is a time-independent induction signal following the pulse. We assume that the effect of the existence over the sample of differing ∇E (due to crystalline imperfections) or internal magnetic fields (due to dipolar interactions) is to dephase the precessing microscopic magnetization vectors and to cause the induction signals to decay in a time T_2 . Figure 1 is a multiple exposure of decay signals in a NaClO_3 single crystal taken with and without an H_0 . The decay envelope is Gaussian with T_2 equal to 425 μsec . A calculated T_2 of this order of magnitude results from a computation of the magnetic dipolar interaction of a Cl nucleus with the nuclear moments located within one lattice parameter.

To compute the Zeeman effects, we consider an H_0 applied at an angle θ_0 with respect to ∇E and an H_1 applied for a time t_w at an angle θ_1 . The voltage induced in the coil for $t > t_w$ is then given by:

$$V(t) \propto \sin\theta_1 \sin(\sqrt{3}\Omega_1 t_w \sin\theta_1) \left[\frac{f-1}{f} \cos\left\{ \frac{\Omega_0(3+f) \cos\theta_0}{2} t \right\} + \frac{f+1}{f} \cos\left\{ \frac{\Omega_0(3-f) \cos\theta_0}{2} t \right\} \right], \quad (1)$$

where $\Omega_0 = \gamma H_0$, $\Omega_1 = \gamma H_1$, and $f = (1 + 4 \tan^2 \theta_0)^{\frac{1}{2}}$. The appearance, in general, of the two frequencies in square brackets in (1) arises from the mixing (in the presence of an H_0) of the $\frac{1}{2}$ and $-\frac{1}{2}$ m states, which splits the steady state resonance symmetrically into two pairs of lines.² Further calculations predict a spin-echo signal at 2τ (following the application of a second rf pulse at time τ). We have observed echoes and stimulated echoes in both the powders and single crystals. Hahn and Herzog (see following letter) report the behavior of the echoes as a function of H_0 and of crystal orientation.

In NaClO_3 , ∇E lies along the symmetry axis of the molecule. The four molecules in a unit cell of the crystal are oriented with their symmetry axes along the body diagonals of the unit cube. [Direction cosines $(1/\sqrt{3}, 1/\sqrt{3}, 1/\sqrt{3}; \text{etc.})$] The beat structure in Fig. 1 is that observed on the decay in a single crystal with H_1 and H_0 both applied along $(0,0,1)$. In this orientation $\cos\theta_0 = 1/\sqrt{3}$ and $f=3$ for each of the four molecules in the unit cell. A single-beat frequency $\sqrt{3}\gamma H_0/2\pi$ is then predicted by Eq. (1) and is found to agree with that observed. Figure 2 shows the more complex pattern observed with H_1 along $(0,0,1)$ and H_0 along $(1/\sqrt{2}, 1/\sqrt{2}, 0)$. The voltage predicted by (1) is here proportional to $2 \cos(\Omega_0 t) + 1.6 \cos(0.53\Omega_0 t) + 0.4 \cos(1.9\Omega_0 t)$, which agrees with the pattern of the observed decay. The echo exhibits a structure similar to that on the decay.

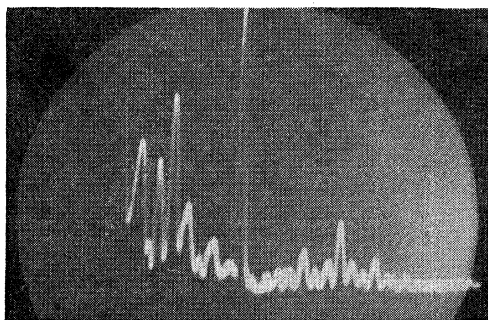


FIG. 2. Induction decay and echo found with H_1 along $(0,0,1)$ and an H_0 of 12 gauss along $(1/\sqrt{2}, 1/\sqrt{2}, 0)$. The separation of the pulses is 900 μsec .

The single crystals of NaClO_3 were loaned by the Bell Telephone Laboratories. We are grateful to E. L. Hahn and B. Herzog for discussions of their results and have also benefited from conversations with C. P. Slichter and D. McCall. Much of our rf equipment was designed by H. W. Knoebel and L. S. Kypta.

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¹ E. L. Hahn, Phys. Rev. **80**, 580 (1950).

² C. Dean, thesis, Harvard University (unpublished); Phys. Rev. **86**, 607 (1952).

Anisotropic Relaxation of Quadrupole Spin Echoes

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BY the pulsed nuclear induction method¹ we have observed the free Larmor precession of Cl^{35} and Cl^{37} nuclear moments, due only to the pure crystalline electric field gradient in single and powdered crystals of NaClO_3 . When a small constant magnetic field H_0 is applied to the single crystal, the echo relaxation time due to spin-spin coupling is modified by the variation of the Zeeman splitting as the magnitude and direction of H_0 normal to the cubic axis of a NaClO_3 single crystal is varied. We refer the reader to the preceding letter by Bloom and Norberg which discusses their independent observations of the free quadrupole precession and theory of the Zeeman splitting.

The formation of nuclear signals following two rf pulses, as shown in Fig. 1, can be explained by the analogous mechanism of spin echo formation in large magnetic fields.¹ The chlorine quadrupole moment Q , with spin $I = \frac{3}{2}$, is aligned by an electric field gradient q which is assumed to be axially symmetric about the molecular bond (z axis) joining Na to Cl. The single quadrupole resonance transition frequency $\omega = eqQ/2\hbar$ is replaced in general by four separate resonance frequencies² as the degeneracy of the $m = \pm \frac{1}{2}$ and $m = \pm \frac{3}{2}$ states is removed by H_0 . In the unit cell there are four chlorine nuclei with their z axes of quantization oriented along the body diagonals of the sub-cubic cells. There are two nonequivalent directions of Na-Cl axes with respect to H_0 , and a pair of chlorine nuclei is assigned to each of these directions denoted by + and -. Using a quantum-mechanical treatment applied in an earlier paper,³ we obtain the solution for free precession in agreement with that given by Bloom and Norberg. The spin echo signal, described by a similar solution, appears symmetric about the time $t = 2\tau$, where τ is the separation between pulses. The observed signal is given by $V = V_+ + V_-$, where

$$V_{\pm} \propto \sin^3(\sqrt{3}\omega_1 t_w \sin\theta_{\pm}) \cos(\sqrt{3}\omega_1 t_w \sin\theta_{\pm}) \times \left\{ \left(\frac{\beta_{\pm} - 1}{\beta_{\pm}} \right) \cos[(\omega_0 \cos\theta_{\pm})(3 + \beta_{\pm})(t - 2\tau)/2] + \left(\frac{\beta_{\pm} + 1}{\beta_{\pm}} \right) \cos[(\omega_0 \cos\theta_{\pm})(3 - \beta_{\pm})(t - 2\tau)/2] \right\} \times \exp\left\{ -[(t - 2\tau)^2/T_2^2 + t^2/(T_2')^2(H_0, \theta_{\pm})] \right\}.$$

θ is the angle between H_0 and the molecular axis, $\omega_0 = \gamma H_0$, $\omega_1 = \gamma H_1$, $\beta_{\pm} = (1 + 4 \tan^2 \theta_{\pm})^{\frac{1}{2}}$, γ is the gyromagnetic ratio, and H_1 is the rf field. In terms of the angle φ which H_0 makes with respect to the 001 direction, $\cos\theta_{\pm} = \sqrt{\frac{3}{2}} \cos(\varphi \pm \pi/4)$ and $\sin\theta_{\pm} = [1 - \frac{3}{2} \cos^2(\varphi \pm \pi/4)]^{\frac{1}{2}}$. We infer from our observations that T_2' for each of the + and - groups is nearly the same. The total width of the echo at half-maximum is given by T_2 seconds (0.5 millisecond for Cl^{35}), which is equivalent to a static local magnetic dipole field inhomogeneity of $\Delta H \approx 2.6$ gauss for an assumed Gaussian distribution. For each setting of τ , with the spins initially at thermal equilibrium, the maximum of the echo amplitude is observed to be proportional to $\exp[-(2\tau)^2/T_2'^2(H_0, \varphi)]$

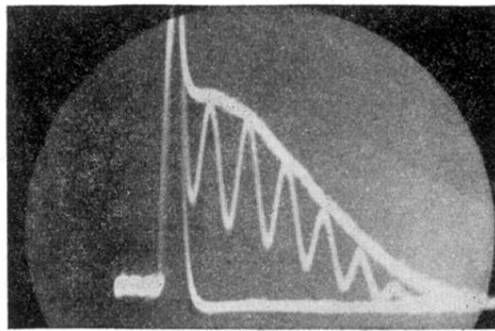


FIG. 1. Induction decay in a single crystal of NaClO_3 . The sweep is $800 \mu\text{sec}$ long. The beat structure appears in the presence of a Zeeman field of 12 gauss applied, parallel to H_1 , along $(0,0,1)$.

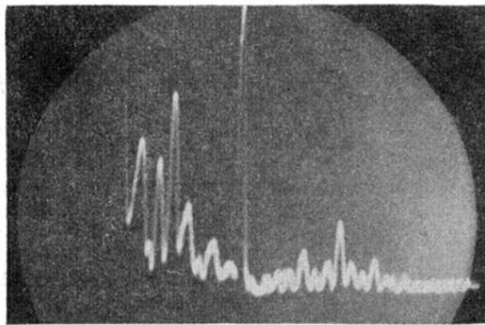


FIG. 2. Induction decay and echo found with H_1 along $(0,0,1)$ and an H_0 of 12 gauss along $(1/\sqrt{2}, 1/\sqrt{2}, 0)$. The separation of the pulses is 900 μsec .