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ION STORAGE TECHNIQUE IN RF-SPECTROSCOPY
(RADIOFREQUENCY RESONANCE REORIENTATION)

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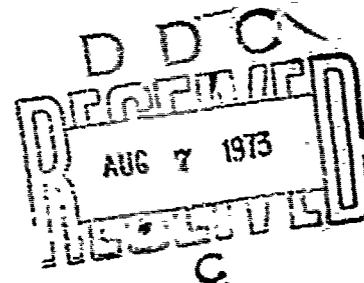
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13. ABSTRACT The results of a program of research in the rf spectroscopy of stored atomic and molecular ions are described. Work has been done on the study of magnetic resonance and hfs of e^- , $^4\text{He}^+$, $^3\text{He}^+$ and H_2^+ . Techniques for measuring the temperature of ion gases and their radiative cooling have been developed. Extension of the techniques developed under the grants resulted in the highest resolution for any atomic line ($^{199}\text{Hg}^+$ hfs) ever achieved.			

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14. KEY WORDS	LINK A		LINK B		LINK C	
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Atomic Physics Molecular Physics Ion-Storage Collision Technique in rf Spectroscopy Magnetic Resonance Hyper Fine Structure Ions: $^4\text{He}^+$, $^3\text{He}^+$, H_2^+ "Bolometric" Technique Ion Gas Cyclotron-resonance e highest resolution in an atomic line						

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The results of the work performed under the partial sponsorship of the grants are described in the 29 publications tabulated in the appended list. Also a brief survey of this work prepared by the principal investigator, "rf-Spectroscopy of Stored Ions" reprinted from Atomic Physics, Plenum Press 1969, is attached. Further, a detailed review article "Radiofrequency-Spectroscopy of Stored Ions", Advances in Atomic Physics 3, 53 (1967) and 5, 109 (1959) is available, of which the table of contents is appended.

It is a pleasure to note the group of younger men who have participated in the work of the contract; these include postdoctoral men brought in from other institutions, men who had been awarded the Ph.D. degree at the University of Washington and remained to follow promising lines of research and to capitalize on an investment in equipment and on their immediate facility with the body of ideas with which the work of the grants is concerned, and those having earned doctorates.

The list of publications records the support by the subject grants of work of the following postdoctoral men who had earned the Ph.D. degree at institutions other than the University of Washington:

E.N. Fortson (Ph.D. Harvard, 1962)
now Associate Professor of Physics, University of Washington

H.R. Feldman (Ph.D. Columbia, 1963)
now with Applied Physics Laboratory, University of Washington

C.B. Richardson (Ph.D. U. of Pittsburgh, 1962)
now Assistant Professor of Physics, University of Arkansas

H.A. Schuessler (Ph.D. Heidelberg, 1964)
now Associate Professor of Physics, Texas A&M University

G.H. McCall (Ph.D. Princeton, 1967)
now with A.E.C.

Talbert Stein (Ph.D. Brandeis, 1967)
now Assistant Professor, Wayne State University

D. Wineland (Ph.D. Harvard, 1970)
now Postdoctoral Associate, University of Washington

R. Van Dyck (Ph.D. Berkeley, 1970)
now Postdoctoral Associate, University of Washington

The following have earned doctoral degrees and when starred have continued for varying periods as postdoctoral associates at the University of Washington:

E.S. Ensberg, thesis (1962): Experimental Upper Limit for the Permanent Electric Dipole Moment of Rb^{85} by Optical-Pumping Techniques
now with Physics Section, Bell Telephone Laboratories

F.G. Major*, thesis (1962): The Orientation of Electrodynamically Confined He^+ Ions
now with NASA

K.B. Jefferts*, thesis (1962): Alignment of Trapped H_2^+ Molecular Ions By Selective Photodissociation
now with Physics Section, Bell Telephone Laboratories

D.A. Church, thesis (1969): Storage and Radiative Cooling of Light Ion Gases in Radiofrequency Quadrupole Traps
now Postdoctoral Associate, Berkeley

P.A. Ekstrom*, thesis (1971): Search for a Differential Stark Shift of the Cs- and Rb- Magnetic Resonance Frequencies Employing Atomic Light Modulation Oscillators
now Postdoctoral Associate, University of Washington

S.C. Menasian*, thesis (1973): High Resolution Study of the $(F_2)=(3/2, 1/2) + (1/2, 1/2)$ hfs Transition in Stored H_2^+ Molecular Ions
now Postdoctoral Associate, University of Washington

F.L. Walls, thesis (1970): Determination of the Anomalous Magnetic Moment of the Free Electron from Measurements on an Electron Gas at 20^oK Using a Bolometric Technique
now Postdoctoral Associate, JILA

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Numbers

PUBLICATIONS

- 1 Spin Resonance of Free Electrons Polarized by Exchange Collisions, H. G. Dehmelt, Phys. Rev. 109, 381 (1958); Bulletin APS 3, 369 (1957) (T) (invited paper)
- 2 Preservation of Spin State in Free Atom-Inert Surface Collisions, ROBINSON, ENSBERG, and DEHMELT, Bulletin APS 3, 9 (1958) (A)
- 3 Optical Transmission Monitoring of Free Atom Resonance, H. G. Dehmelt, Proceedings Duke Radio and Microwave Spectroscopy Conference, November 4-6, 1957.
- 4 Realization and Measurement of Long Free Atom Spin State Life Times, H. G. Dehmelt, Proceeding USASEL Frequency Control Symposium, May 6-8, 1958.
- 5 Spin Resonance of Free Electrons, H. G. Dehmelt, Proceedings CNRS Symposium, Paris, July 8-9, 1958; Journal de Physique et le Radium 19, 866 (1958).
- 6 Spin Exchange Resonance of Free Electrons, H. G. Dehmelt, abstract of an invited paper given at the Ann Arbor Conference on Optical Pumping, June 15, 1959.
- 7 Polarization of Atomic Hydrogen at Low Pressures, F. G. Major, unpublished
- 8 Alignment of the H_2^+ Molecular Ion by Selective Photodissociation I, H. G. Dehmelt and K. B. Jefferts, Phys. Rev. 125, 1318
- 9 Orientation of $(He^4)^+$ Ions by Exchange Collisions with Cesium Atoms, H. G. Dehmelt and F. G. Major, Phys. Rev. Letters 8, 213 (1962)
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- 11 The Orientation of Electrodynamically Contained $(He^3)^+$ Ions, F. G. Major and H. G. Dehmelt, Bulletin A.P.S. 7, 432 (1962)
- 12 HFS of $(He^3)^+$ Ground State by an Ion-Storage Exchange-Collision Technique, E. N. Fortson, F. G. Major, and H. G. Dehmelt, Bulletin A.P.S. 9, 626 (1964)
- 13 Ultrahigh Resolution $(He^3)^+$ HFS Spectra by an Ion-Storage Collision Technique, E. N. Fortson, F. G. Major, and H. G. Dehmelt, Phys. Rev. Letters 16, 221 (1966)

- 14 Experimental Upper Limit for the Permanent Electric Dipole Moment of Rb^{85} by Optical-Pumping Techniques, E.S. Enskog, Phys. Rev. 153, 36 (1967)
- 15 Radiofrequency Spectroscopy of Stored Ions I: Storage, H. G. Dehmelt, Academic Press Inc, New York, 1967, Adv. in Atomic and Molecular Physics, Vol. 3
- 16 Alignment of the H_2^+ Molecular Ion by Selective Photo-dissociation, II. Experiments on the Radio-Frequency Spectrum*, C. B. Richardson, K. B. Jefferts, and H. G. Dehmelt, Phys. Rev. 165, 30 (1968)
- 17 Magnetic Resonance Spectrum of the He^3 Ion by an Ultra-high-Resolution Ion-Trapping Exchange-Collision Technique*, Hans A. Schuessler, Stephen C. Menasian, and Norval Fortson, Bulletin A.P.S. 13, 67 (1968)
- 18 Exchange-Collision Technique for the rf Spectroscopy of Stored Ions*, F. G. Majori and H. G. Dehmelt, Phys. Rev. 170 (1968)
- 19 rf-Spectroscopy of Stored Ions, H. G. Dehmelt, (invited paper), International Conference on Atomic Physics, Victor W. Cohen, Gisbert zu Putlitz, New York University, June 3-7, 1968
- 20 Collisional Orientation of the Molecular H_2^+ Ion and Observation of Transitions Between the Zeeman Levels of the Rotational Ground State*, H. A. Schuessler and H. G. Dehmelt, International Conference on Atomic Physics, Gisbert zu Putlitz, Victor W. Cohen, New York University, June 3-7, 1968
- 21 "Bolometric" Technique for the rf Spectroscopy of Stored Ions*, H. G. Dehmelt and F. L. Walls, Phys. Rev. 21, 127 (1968)
- 22 Physics of the One-and-Two-Electron Atoms, HFS Spectrum of $^3\text{He}^+$ by the Ion Storage Collision Technique, H. G. Dehmelt, (invited paper) Sommerfeld Centennial Memorial Meeting, Munich, 9-14 September 1968
- 23 Zeeman Splitting of Stored Molecular H_2^+ and Atomic He^+ Ions*, Hans A. Schuessler, Bulletin A.P.S. 13, 1674 (1968)³
- 24 Radiofrequency Spectroscopy of Stored Ions II: Spectroscopy, H. G. Dehmelt, Academic Press Inc., New York, 1969, Adv. in Atomic and Molecular Physics, Vol. 5
- 25 Storage Ring Ion Trap Derived from the Linear Quadrupole Radio-Frequency Mass Filter, D. A. Church, J. Appl. Phys. 40, 3127 (1969)
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- 27 Hyperfine Structure of the Ground State of $^3\text{He}^+$ by
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E. H. Fortson, and H. G. Dehmelt, Phys. Rev. 187, No. 1 (1969)
- 28 Differential Linear Stark Shift in Rb^{85} and Cs^{133} ,
P. A. Ekstrom, Bulletin A.P.S. 16, 849 (1971)
- 29 High Resolution Study of the $(1, 1/2, 1/2) - (1, 1/2, 3/2)$
hfs Transition in H_2^+ , S. C. Menasian and H. G. Dehmelt, Bulletin
A.P.S. 18, 408

Extension of our techniques by F. G. Major, formerly associated with
the grants, and a German coworker, has resulted in the highest resolution
for any atomic line ever achieved, namely 10 Hz out of 4×10^{10} Hz for the
 $^{199}\text{Hg}^+$ hfs; [F. G. Major and G. Werth, Phys. Rev. Letters 30, 1155 (1973)].
Simultaneously, a resolution of 6×10^3 Hz out of 10^{14} Hz has been reported
for the $^{12}\text{CH}_4$ molecule by laser saturated absorptive spectroscopy [S. L.
Hall and C. Borde, Phys. Rev. Letters 30, 1101 (1973)].

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rf-SPECTROSCOPY OF STORED IONS

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Experimental attempts to approach the ideal of isolated atomic systems floating at rest in free space for unlimited periods and free from any undesired outside perturbations appear to be worthwhile for a variety of reasons. At the same time such experiments are of limited value unless one also devises means for first preparing the atomic systems in certain selected states and for later observing the development of these states in time due to internal or controlled external interactions, as in hfs or magnetic resonance experiment. The first experimental goal therefore is to develop techniques to isolate, contain in a trap, thermalize, and possibly refrigerate the atomic systems under study. Since these problems appear to be most easily solved for charged particles, we restrict ourselves in the following to ions. Next, collision reactions with suitable, state-selected projectiles may serve to create oriented, aligned, or somehow state-selected ions from atoms or cause ions already present in the trap to undergo transitions to selected states. A second reaction, not necessarily of the same type, may be used for analysis or interrogation of the stored ions. A whole arsenal of suitable projectiles becomes available once ion containment times of sufficient duration, of the order of seconds, are realized, bringing down the necessary projectile flux densities to feasible values.

General discussions of the subject have been given by the author who also previously suggested rf-spectroscopy of stored ions (Dehmelt, 1956a, 1962, 1963, 1967). An experiment (Dehmelt and Major, 1962; Major, 1962; Major and Dehmelt, 1968) carried out on He^+ ions with the intent of observing their magnetic resonance may serve as an illustrative example; He^+ ions contained under ultra-high vacuum conditions in an electric quadrupole rf trap (Paul et al.,

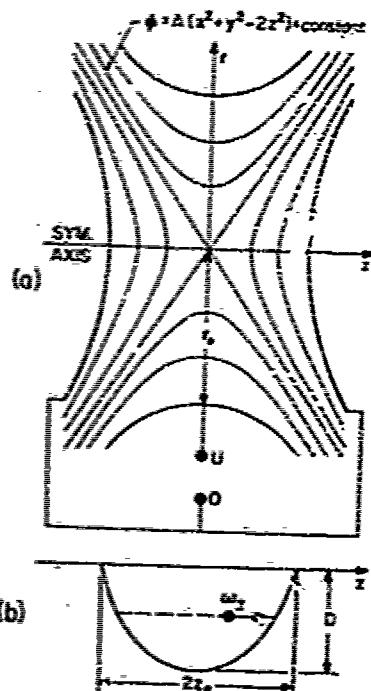


Fig. 1. Hyperbolic electrode configuration employed in ion storage devices useful in rf-spectroscopy. Application of an alternating voltage $U = V \cos \Omega t$ at the terminals shown in (a) creates a three-dimensional harmonic oscillator potential of depth $D = D_0$ along the z axis as depicted in (b). When a dc voltage $U = U_0$ is applied the potential at the origin has a saddle point only. While a parabolic well is also obtained along the x axis, motion of the charged particles in the z -direction has to be restricted in this embodiment of the Penning discharge geometry by a strong axial magnetic field as usual. The traps may be filled by electron impact ionization of the residual gas inside them.

1958; Fischer, 1959; Wuerker et al., 1959) were bombarded with polarized Cs atoms. In the ensuing spin-exchange collisions, the He^+ electron spins became oriented according to the reactions



Fig. 3

while the Cs atoms lost orientation. This experiment is closely related to an earlier experiment (Dehmelt, 1956b, 1958a, b; Balling and Pipkin, 1965) in which free electrons contained by a positive ion cloud were oriented by spin-exchange collision with oriented sodium atoms,



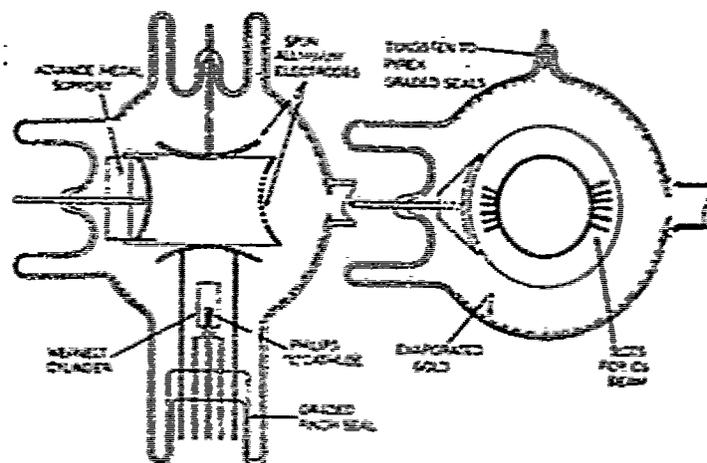


Fig. 2. Constructional details of the quadrupole-ion cage assembly used in Majka's experiments. Operating parameters are given in Table I.

In both instances, magnetic resonance disorientation of the ions He^+ and Li^+ , the process of interest, was monitored by a second collision reaction. In general, information about the magnetic resonance is contained in the final states of all the reaction products. In the electron experiment the monitoring focussed on the orientation loss of the sodium atoms or of the "projectiles". Since spin-exchange reactions between oriented electrons e^{\uparrow} and Na^{\uparrow} atoms do not produce Na^{\uparrow} atoms while $e^{\uparrow} + \text{Na}^{\uparrow}$ collisions do, the appearance of Na^{\uparrow}

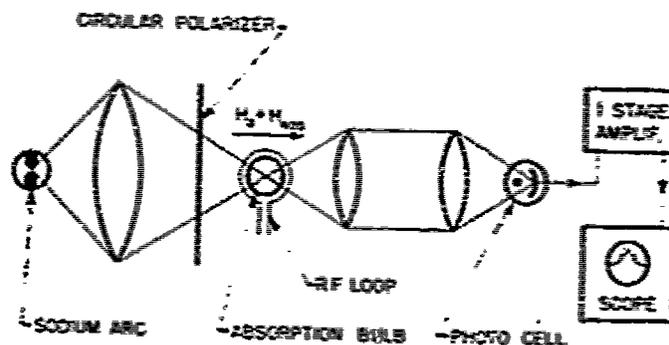


Fig. 3. Apparatus for observation of spin resonance of free electrons polarized by spin exchange collisions. The absorption bulb contained a cold plasma and Na-atoms at densities of $N \sim 10^8 \text{ cm}^{-3}$ and $n \sim 10^{10} \text{ cm}^{-3}$ respectively diffusing in an Argon buffer gas at ~ 30 Torr. The orientation of the Na-atoms was created and monitored by circularly polarized resonance light.

TABLE I

DIMENSIONS, OPERATING PARAMETERS, OBSERVED AND
DERIVED ION DATA FOR ILLUSTRATIVE TRAP

Axial dimension:	$z_0 = 2.5$ cm
Radial dimension:	$r_0 = 3.5$ cm
Effective trap volume:	$V_t = 128$ cm ³
dc bias:	$U_0 = 7$ volt
ac drive amplitude:	$V_0 = 175$ volts
Frequency:	$\Omega = 2\pi \times 1$ MHz
Vacuum:	$p \approx 3 \times 10^{-8}$ Torr
Background:	Mostly H ⁺
Electron current:	$i_e = 1$ mA
Electron acceleration voltage:	$U_e = 400$ volts
Electron pulse duration:	~ 0.1 sec
Ionic species:	[He ⁴⁺] ⁺
Axial oscillation frequency:	$\omega_z = 2\pi \times 110$ kHz
Maximum velocity in trap center:	$z_0 \omega_z = 1.73 \times 10^6$ cm/sec
Axial depth:	$D_z = 6.2$ volts
Radial depth:	$D_r = 8.2$ volts
Maximum instantaneous energy:	$W_{max} \approx 12.4$ eV
Maximum experimental stored charge:	$q \approx 10^7 e$
Maximum stored charge:	$q_{max} \approx 3 \times 10^8 e$
Ion lifetime:	$T = 8$ sec
(maximum allowed):	$T = 50$ sec
Self-collision parameter:	$T_c^* = 1.5$ sec
He ⁺ -He charge exchange:	$T_{ce} = 0.3$ sec

did serve as an indicator for the magnetic resonance disorientation of the electrons. In the He⁺ experiments the orientation monitoring phase focussed upon the He⁺ targets and not the Cs⁺ projectiles. By employing the nearly resonant charge-exchange reaction leading to excited He states,

Fig. 4

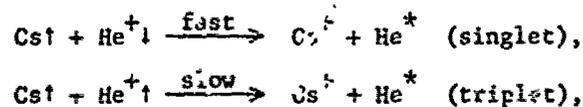


Fig. 5 it was possible to translate the magnetic resonance disorientation of the initially oriented He⁺ ions to He⁺ into a reduction of their

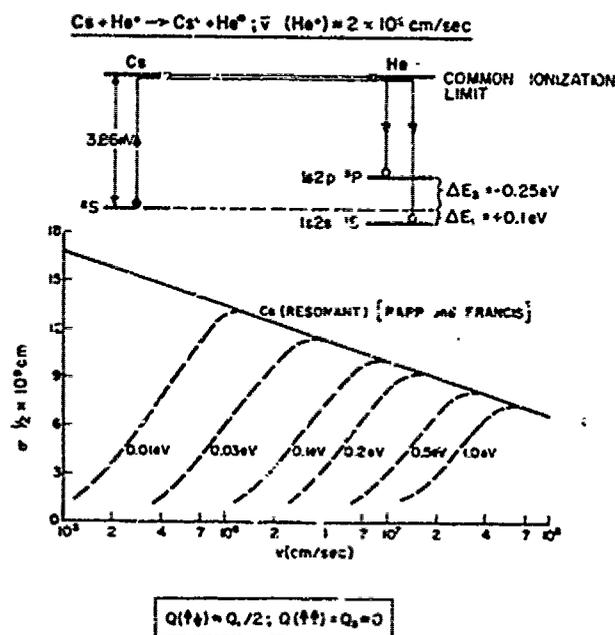


Fig. 4. Theoretical curves of Rapp and Francis (1962) for charge-exchange cross section Q or σ as a function of the velocity in near-resonant collisions involving Cs, with the energy defect ΔE , as the single essential parameter, and energy levels important in the $Cs + He^+$ reaction.

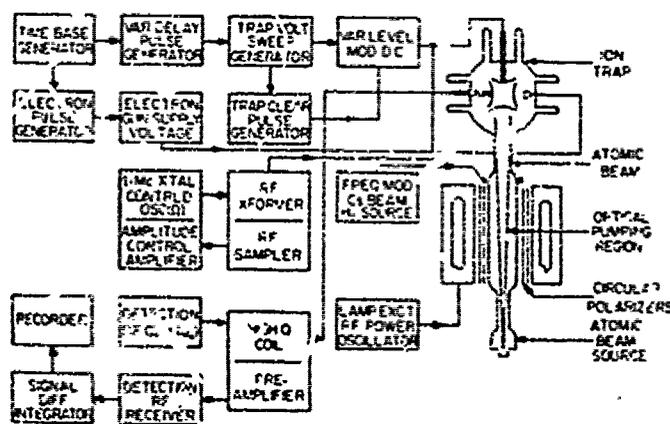
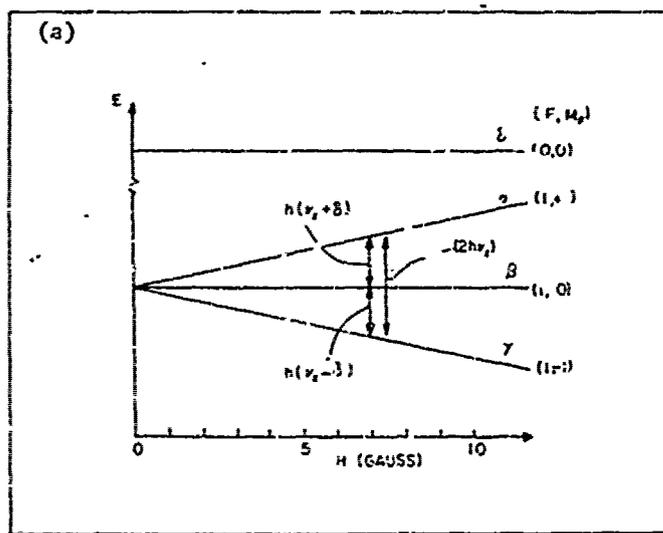


Fig. 5. Block diagram of apparatus for observation of orientation and magnetic resonance of $\sim 10^7$ stored He^+ ions. Large Helmholtz coils provided the constant magnetic field H_0 , horizontal and in the plane of the drawing.



(b)

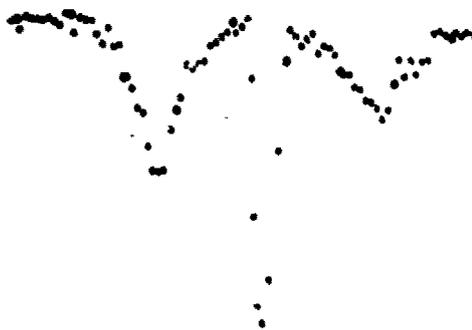
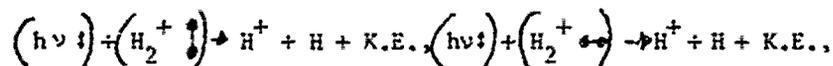


Fig. 6. (a) Low-field region of Breit-Rabi diagram for the hfs energy levels of $(\text{He}^3)^+$ as a function of magnetic field H . (b) Digital-analyzer display of the rf saturated $\Delta F=0$ spectrum of $(\text{He}^3)^+$ obtained with the Cs optically pumped into the $m_s = -1/2$ state resulting in a depolarization signal S' greater for the lower frequency $(1, -1)$ - $(1, 0)$ transition than the $(1, 0)$ - $(1, +1)$ transition. The relatively narrow line in the middle is the double quantum transition $(1, -1)$ - $(1, +1)$. The magnetic field had the value $H=7.23$ G for which the spectrum was centered about 10 MHz and the Paschen-Back-Coudermit shift amounted to $\delta = 11.5$ kHz. The frequency increases from left to right, and the quantity plotted is simply $n(0.8 \text{ sec})$, the number of stored ions remaining after the interaction interval, averaged over 80 traversals of the spectrum with the zero strongly suppressed.

- Fig.6 number, which in turn could be conveniently measured. The magnetic resonance of the rotational groundstate of H_2^+ has also been observed with the same apparatus in an analogous experiment (Schuessler and Dehmelt, 1968). The appearance of a new reaction product, namely, Cs^+ , might have provided an even more effective indicator of the He^+ resonance. In fact, in a third experiment on H_2^+ ions (Dehmelt and Jefferts, 1962; Jefferts, 1962; Richardson et al., 1968), using linearly polarized photons as projectiles in the dissociation reaction



the appearance of H^+ was used to monitor the magnetic resonance of H_2^+ . This again is possible because the photo-dissociation rate depends on the angle between electric light vector \uparrow and the internuclear axis \uparrow . As illustrated by the three experiments just described, our studies seem to indicate that a wide variety of target-projectile combinations of interest may be imagined. The state selection of the projectiles necessary may be purely by energy eigenstate, as low as that found in an unpolarized monoenergetic parallel beam of electrons (alignment by electron impact), or as high as that in a beam of circular polarized optical resonance radiation (optical pumping), or in a beam of polarized alkali atoms (polarization by spin exchange).

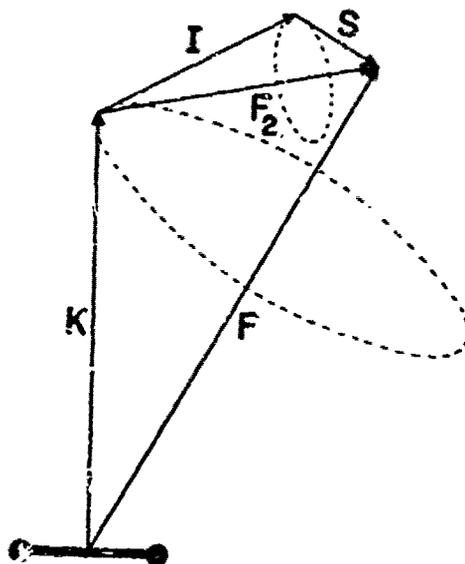


Fig. 7. Coupling scheme of the angular momentum vectors in the H_2^+ ion.

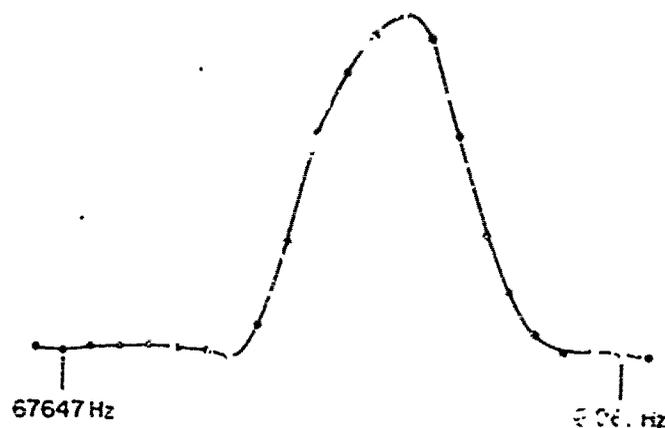


Fig. 8. Unresolved magnetic resonance signal of the (KF_{2f}) -states $(1\ 3/2\ 5/2)$, $(2\ 1/2\ 3/2)$, $(2\ 1/2\ 5/2)$, all having $g_F \approx 2/5$, of the H_2^+ ion in a field $H_0 \approx 115$ G. A point-by-point digital analyzer display obtained by Richardson in 11 hours is shown.

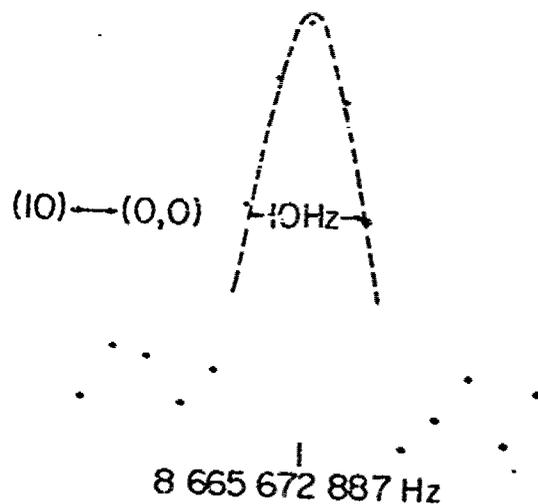


Fig. 9. Digital analyzer display of the $(0,0)$ - $(1,0)$ hfs transition for $(4e^3)_f$ obtained by Schuessler by means of a scheme employing sequential transition pulses at Zeeman- and hfs-frequencies. Recording time was 8 hours.

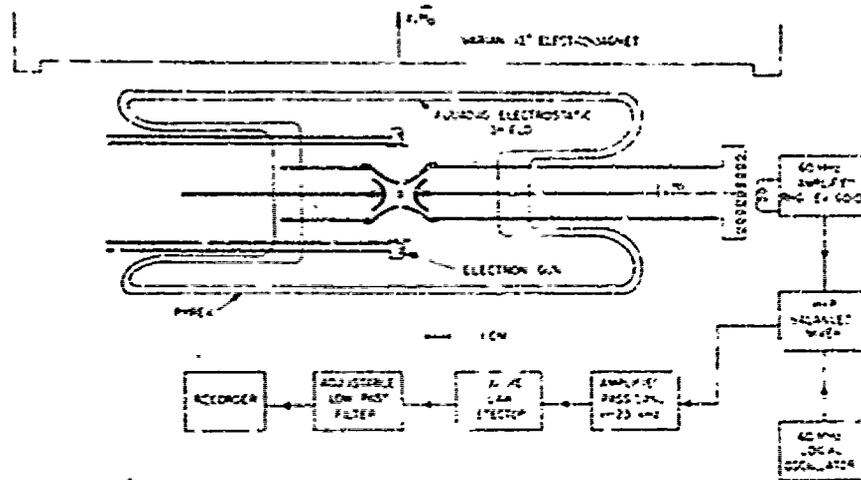


Fig. 11. A cross sectional view of the electron trap and a block diagram of the electronics used in the bolometric detection of the electron cyclotron resonance. The small rectangular slot in the center "ring" electrode is a microwave window. The vacuum manifold with its 15 μ /s Vacion pump and GE 22Gt 210 Triggered Discharge Gauge is connected to the front section of the glass envelope which was cut away to expose this view.

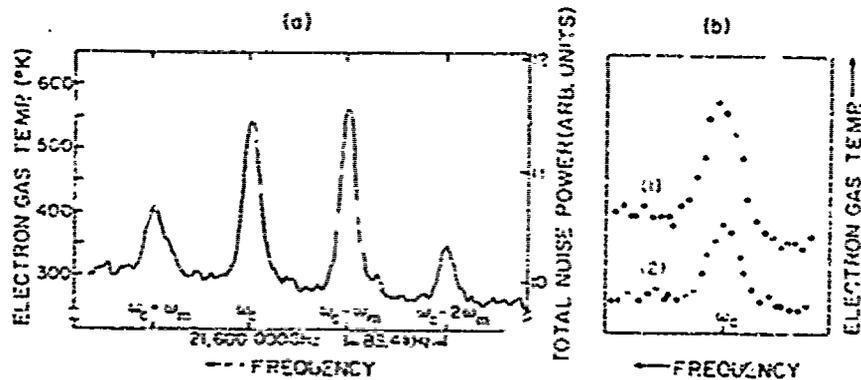


Fig. 12. (a) Temperature increase of the electron gas as the exciting frequency is swept through the cyclotron resonance. The temperature scale is correct to approximately 50°K. The slight slope of the base line is due to losses from the initially injected sample of $\sim 10^5$ electrons during the duration of the sweep which was approximately 4 minutes. (b) The cyclotron line with (1) and without (2) parametric post-multiplication of the electron gas temperature.

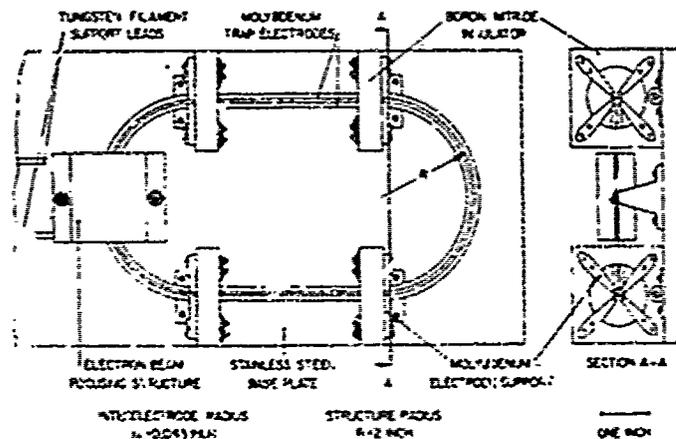


Fig. 13. Race track shaped 4-wire rf quadrupole storage ring constructed by Church. The straight sections of this ring make it well suited for the study of interactions of the stored ions with beams of particles or photons.

absorption of the Hg^+ resonance radiation by the stored ions.

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RADIOFREQUENCY SPECTROSCOPY OF STORED IONS I: STORAGE*

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Seattle, Washington

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SPECIAL TECHNICAL REPORT

July 9, 1973

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but carried out mostly with funds from other sources

Continuous Electronic Observation
of Single Elastically Bound Electron*
(Monoelectron Oscillator)

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U. of Washington

The electron and its antiparticle, the positron, are the most common, most important and the theoretically best understood of the elementary particles. Experimental studies of their properties aiming at improved accuracies consequently do not need much justification. In the present work we are concerned with experimental techniques to cage single electrons and positrons in ultrahigh vacuum by suitable force fields so that they may be subjected to extended observation - weeks have been realized. Since a single electron may be isolated and stored, perturbing interactions with like particles are eliminated. Collisions with the residual gas occur only about every 10 sec because of the ultrahigh vacuum.

The electron is contained in a trap, the principle of which is due to Penning (1936). The trap is formed, Fig. 1, by two cap electrodes C and a ring electrode RE mounted in an evacuated Pyrex envelope P, held between the pole pieces of an electromagnet M generating a field of 4-8 KGauss. The trap structure is axially symmetric about the (horizontal) magnetic field direction and an electron on the axis sees a parabolic potential well of depth

$V_0/2$, V_0 the applied electric trapping potential. An electron placed in this well will carry out a nearly harmonic oscillation at $\nu_2=60$ MHz for $V_0=12$ Volts. Some of the fieldlines emanating from the trapped electron end on charges situated on the cap electrodes. These charges move from one cap to the other through the external circuit when the trapped electron is axially displaced. The resultant rf current now passes through the LC ν_2 resonant circuit of shunt resistance R formed by the coil L and essentially the right cap/ring capacitance C and develops a signal U_S . The LC circuit and R is kept at 80°K and because of the strong coupling to it the electron assumes this temperature in ~ 0.1 sec for $U_D=0$. The electron orbit E is then only 0.2 mm long, corresponding to excitation only by the thermal noise voltage associated with R . When a drive voltage U_D of a frequency ν_2' near ν_2 is applied to the left cap, a driven oscillation of larger amplitude and a signal U_S at ν_2' result which may be detected synchronously.

For the purpose of preparing a single electron the trap was first filled with ~ 10 electrons by shooting a beam of electrons from the gun G through a hole in the right cap and knocking off slow electrons from the residual gas inside the trap. Thereby an easily detected signal U_S resulted (Fig. 2) which, for an appropriately adjusted drive U_D , could be made to decay in discrete steps occurring every ~ 5 min as the drive U_D in conjunction with residual gas collisions boils the electrons successively out of the trap into the caps. The last U_S plateau corresponds to a single electron. For reduced drive amplitudes, the much longer

life times mentioned may be realized.

The experimental task of detecting the small signal U_S in the presence of the many orders of magnitude larger drive U_D was greatly simplified by using one of two alternate schemes: In (A) V_0 is slightly modulated at a frequency $\nu_m \approx 1$ MHz whereby a more easily observable signal at $\nu_z' + \nu_m = \nu_z$ is generated. An analogous scheme is standard praxis in high resolution NMR spectroscopy. In (B) the electron oscillation at $\nu_z' = \nu_z$ is parametrically excited by applying an U_D at $2\nu_z'$.

Similar preparation and detection methods should be applicable to single atomic ions with appropriate design changes. The previous record appears to be held here by Rettinghaus (1967), who, using a bridge circuit, reported a sensitivity sufficiently high to detect four atomic ions.

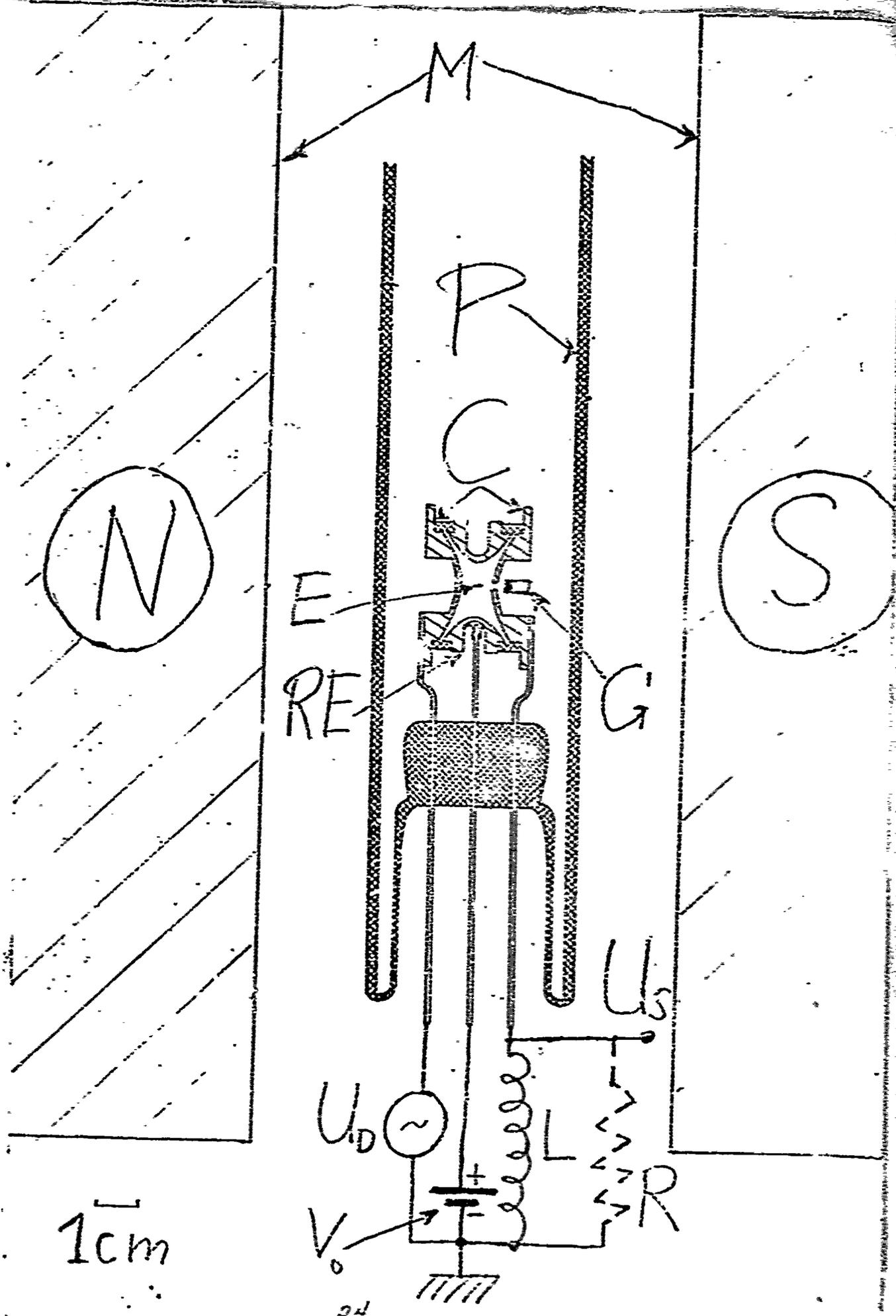
*Previously reported briefly at the East Lansing APS Meeting 1973
Bulletin A.P.S. 18, 785 (1973)

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Rettinghaus, G. (1967) Z. Angew. Phys. 22, 321

Fig. 1. Trapping apparatus for confining, thermalizing and continuously observing a single particle (electron or positron). For explanation of symbols, see text.

Fig. 2. Forced oscillation signal U_S versus time. The signal at $\nu_z = 60$ MHz for an initially injected bunch of electrons decreases discontinuously as the electrons are successively boiled out the trap by the drive U_D . The last plateau is due to a single electron. Detection scheme (A) was used.



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FIG. 1

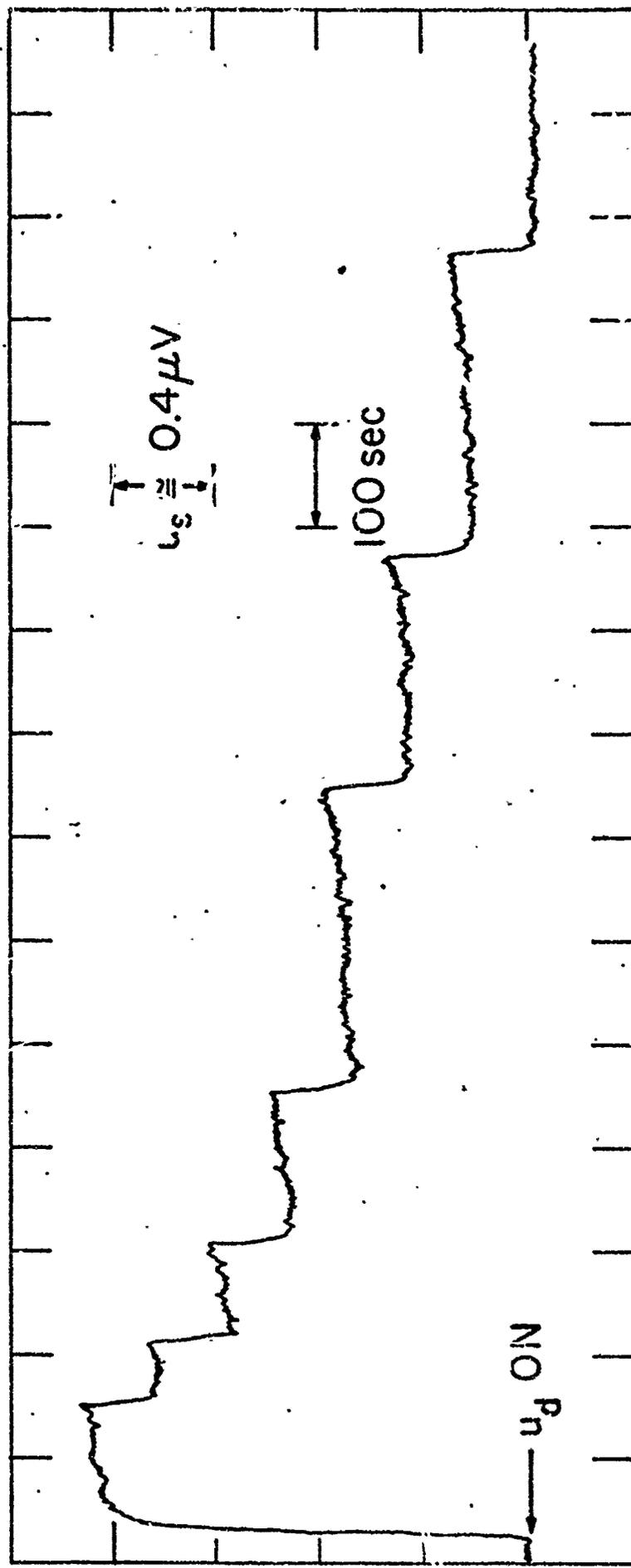


FIG. 2