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EXPERIMENTS ON ELECTRONIC EXCITATION OF ATOMS
PERFORMED AT LAWRENCE RADIATION LABORATORY
BERKELEY, CALIFORNIA, U. S. A.

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"Electronic Excitation of an Atomic Vapor.
Application to Spectroscopy"
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38 - Grenoble, France, May 23-26, 1966

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EXPERIMENTS ON ELECTRONIC EXCITATION OF ATOMS PERFORMED AT LAWRENCE RADIATION LABORATORY, BERKELEY, CALIFORNIA, U. S. A.

Tetsuo Hadeishi

March 8, 1966

EXPERIMENTS ON ELECTRONIC EXCITATION OF ATOMS PERFORMED AT LAWRENCE RADIATION LABORATORY, BERKELEY, CALIFORNIA, U. S. A.*†

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ABSTRACT

A brief survey of the experiment on electronic excitation of atoms, carried on at the Atomic Beam Group of the Lawrence Radiation Laboratory, is made. By means of alignment of excited atoms by electron-impact excitation, we first observed rf paramagnetic-resonance reorientation of Hg and neon metastable states. Later on, the method was extended to observe the coherence property of excited atoms associated with the electron-impact excitation. The coherent excitation of nondegenerate levels of excited atoms results in intensity beats either in the fluorescent decay due to spontaneous radiation or in the resonance absorption by a long-lived metastable state.

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†Work performed under the auspices of the U. S. Atomic Energy Commission.

I. Paramagnetic rf Resonance of Metastable State 3^3P_2 of Ne Produced and Aligned by Electron Impact*

Neon atoms were simultaneously excited and aligned from the $^1S_0$ ground state to the 3^3P_2 metastable state by electron impact. For the low-energy electrons, the magnetic sublevels $M_J = 0$ and $M_J = \pm 1$ were preferentially produced. The alignment property of the metastable state was detected by $\lambda = 6143$ Å resonance absorption by the metastable state. Consideration of spectral-line separation, intensity, and transition probability led us to choose this resonance line. The polarization ratio defined as $p = (\Delta I_\perp - \Delta I_\parallel)/(\Delta I_\perp + \Delta I_\parallel)$—where $\Delta I_\parallel$ and $\Delta I_\perp$ are the absorbed intensity of the resonance radiation whose electric vector is, respectively, parallel and perpendicular to the direction of the electron beam—can be expressed in terms of population of a magnetic sublevel as $(-6N_2 + 3N_1 + 3N_0)/(10N_2 + 7N_1 + 3N_0)$—where N_2, N_1, and N_0 are the population of $M_J = \pm 2$, ± 1, and 0. Therefore, the preferential population of the magnetic sublevels of $M_J = 0$ and ± 1 causes P to be greater than 0. An rf resonance with sufficiently high H_{rf} equalizes the population distribution, which amounts to causing P to equal 0. Figure 1 shows the relative direction of the light, an externally applied magnetic field, and the electron beam. Since the transition probability for $\Delta M_J = 0$, corresponding to the electric vector of resonance radiation parallel to the direction of the electron beam, is 4:1:0 for $M_J = \pm 2$ to ± 2, ± 1 to ± 1, and 0 to 0, the absorption of the resonance radiation at rf resonance increases (Fig. 2).

To observe the rf resonance, we had to obtain a reasonable amount of the metastable-state atoms, and align the metastable state. This was accomplished by adding Hg vapor to a neon-gas-filled diode-structure electron gun which was operated at high current density under space-charge neutralization by Hg^+ ions.[1] By this method we could change the energy of the electron from 11 to 30 V with electron currents as high as 1 A for a 3/4-in. cathode, although normally we operated at a much lower current. Figure 3 shows a cross section of the electron-gun excitation tube. Figure 4 is a block diagram of the experiment. The result of the experiment performed with a neon pressure of about 10 μ is shown in Fig. 5. Magnetic-field full band width of rf resonance was about 100 mG.

Previously we have observed a full bandwidth of about 1 gauss. However, after employing a switch to turn the heater current on and off and observing the signals only during the time in which no current was passing through the heater, we observed the full bandwidth of about 100 mG, indicating a spin relation time of about 10 μsec.

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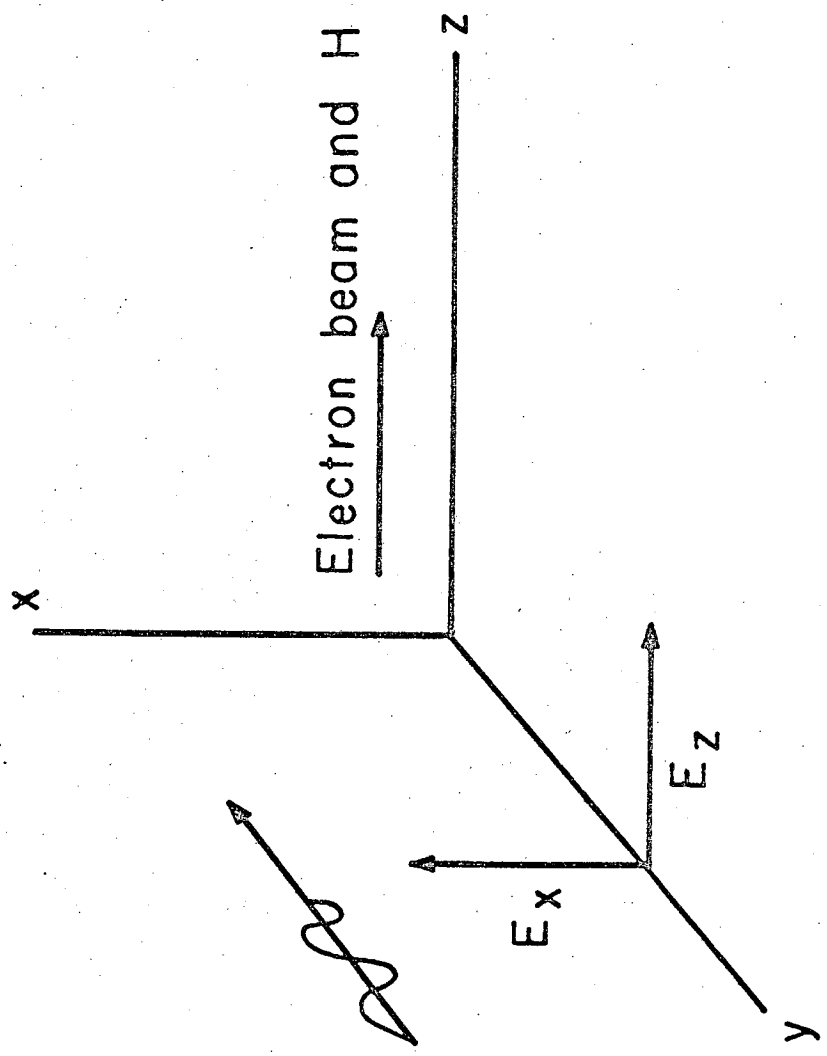
*This was published in Physical Review by T. Hadeishi, O. A. McHarris, and W. A. Nierenberg, Phys. Rev. 138, No. 4, A983 (1965).



MU-28332-A

Fig. 1

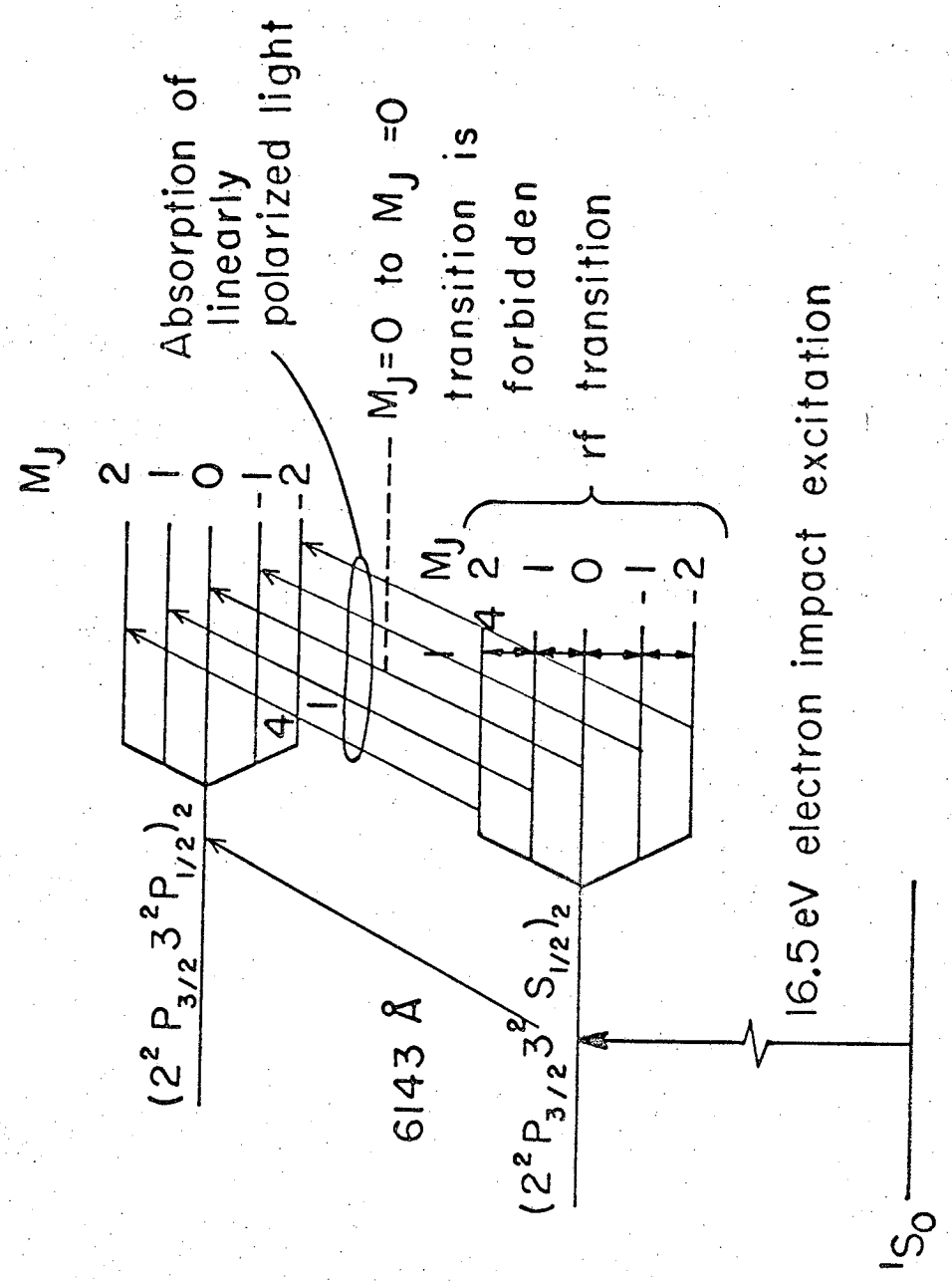


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Fig. 2

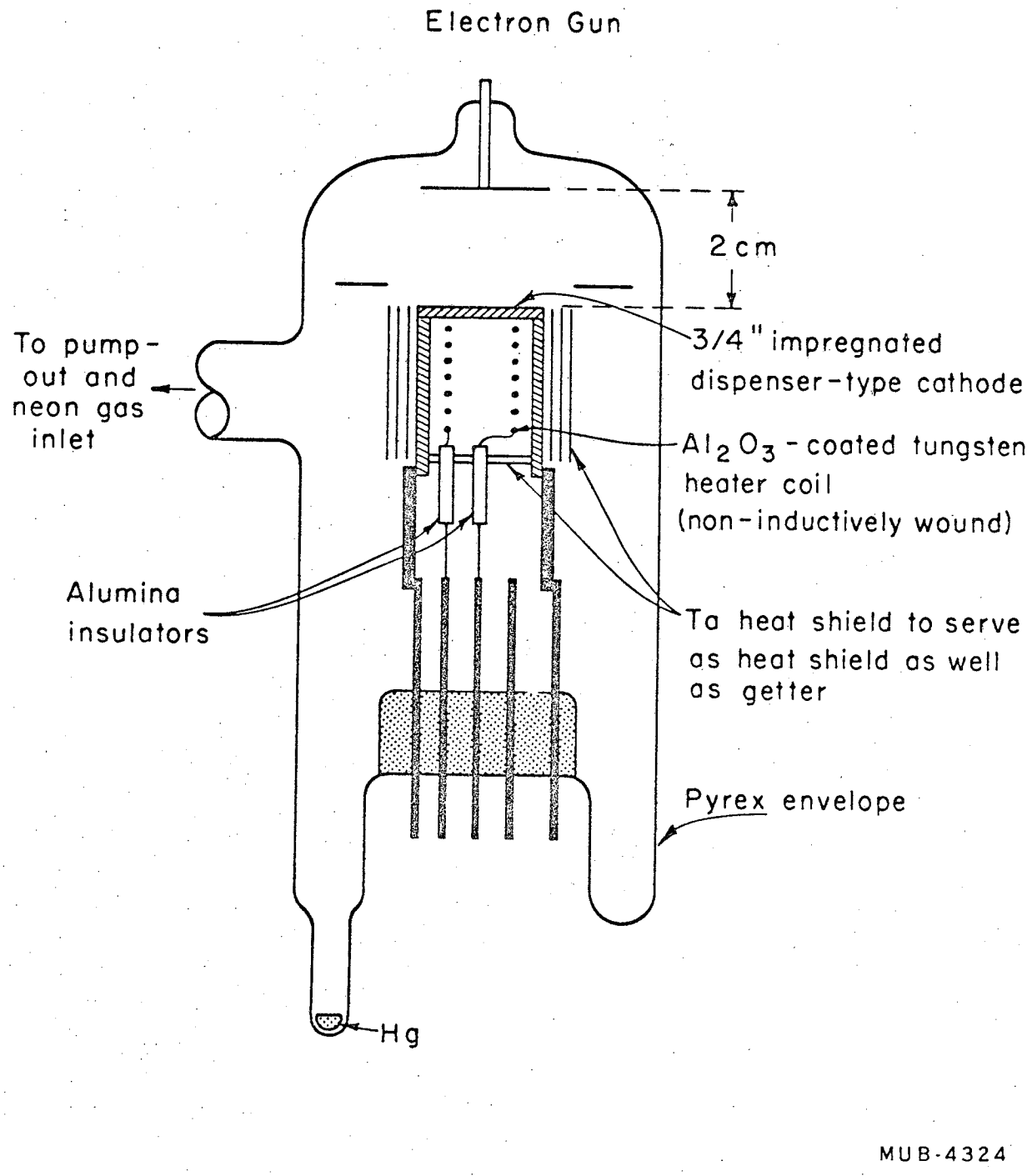
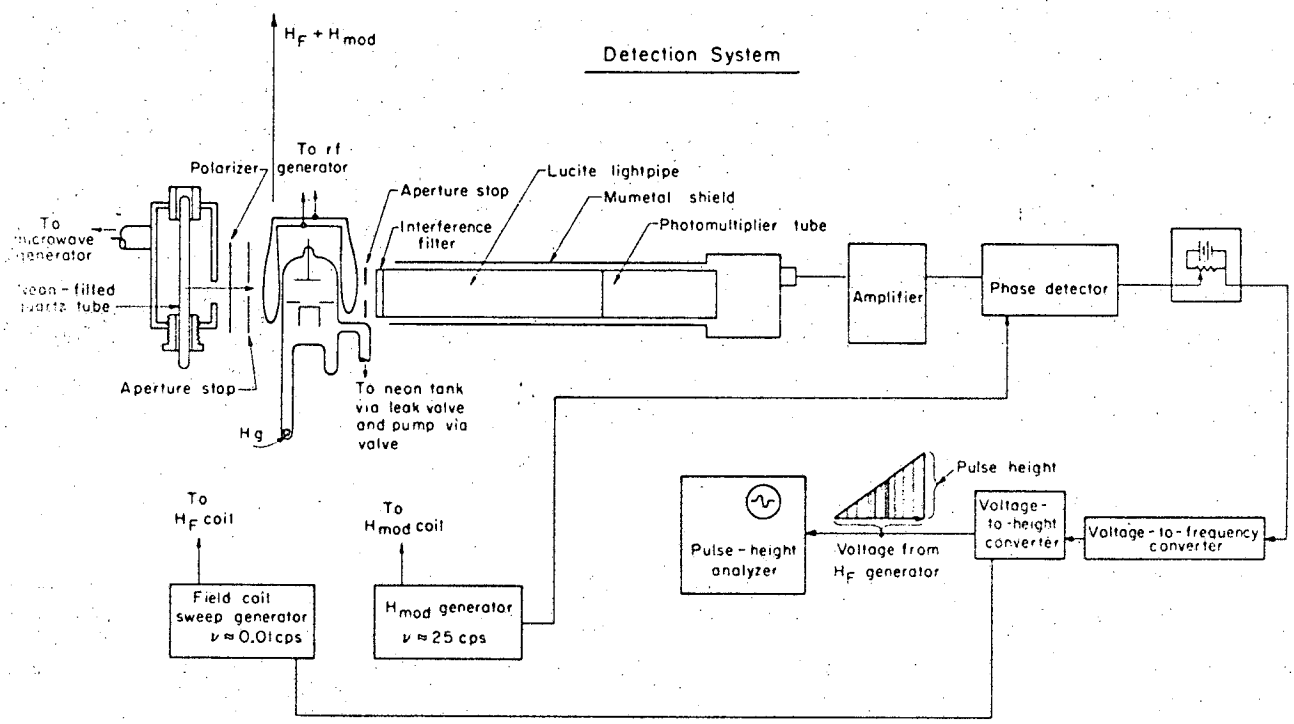

Electron Gun
2 cm
3/4" impregnated dispenser-type cathode
To pump-out and neon gas inlet
Al_2O_3 - coated tungsten heater coil (non-inductively wound)
Alumina insulators
Ta heat shield to serve as heat shield as well as getter
Pyrex envelope
Hg
MUB-4324

Fig. 3



MUB 4335

Fig. 4

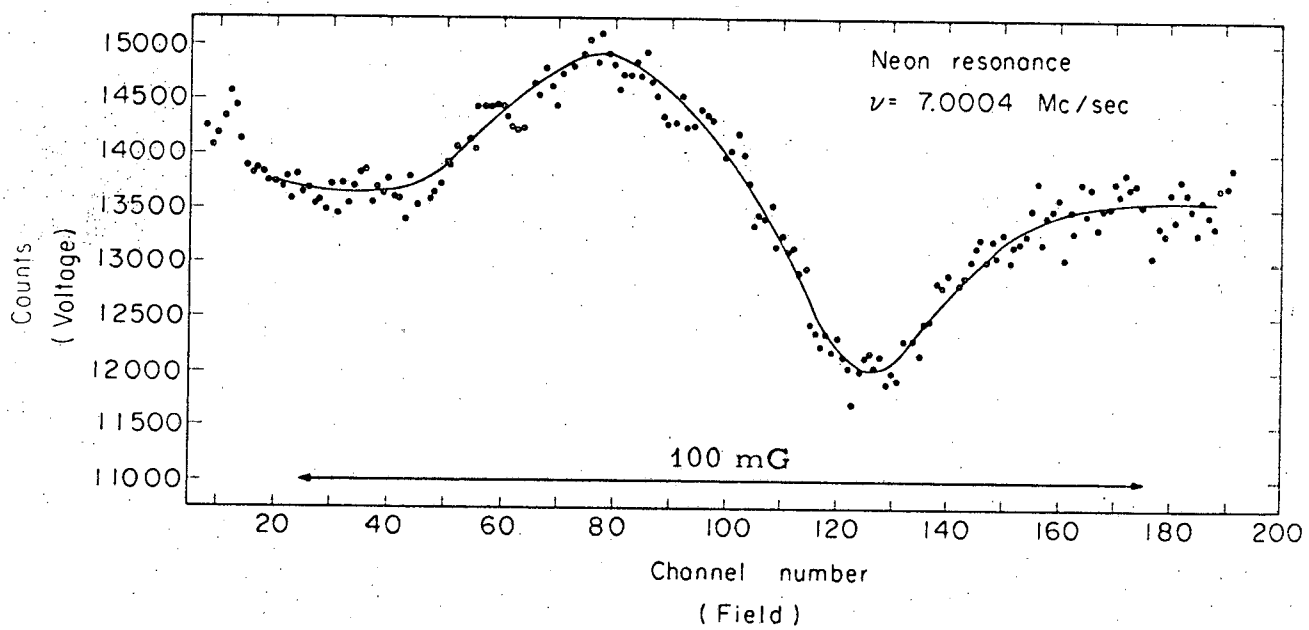

Neon resonance
ν= 7.0004 Mc/sec
100 mG
Counts
(Voltage)
15000
14500
14000
13500
13000
12500
12000
11500
11000
20
40
60
80
100
120
140
160
180
200
Channel number
(Field)

Fig. 5

II. Observation of Intensity Beats Due to Coherent Excitation of Atoms

A. Experiment on 7^3P_1 of Cd by a Pulsed-Electron-Beam Excitation*

We have observed a modulation of exponential decay of resonance luminescence arising from the interference of coherently excited nondegenerate states $M_J = +1$ and -1 of 7^3P_1 of Cd excited by a sharply pulsed electron beam. Similar results were observed by Nedelec, Deschizeaux, and Pebay-Peyroula,[1] and by Aleksandrov[2] by means of modulated-electron-beam excitation.

In the case of Cd whose ground state is 7^1S_0 and whose excited state is 7^3P_1, the $M_J = +1$ and -1 states of 7^3P_1 can be coherently excited when the direction of propagation of the exciting electron is perpendicular to the externally applied magnetic field at an excitation energy slightly above the threshold of excitation. Under such excitation, the probability of photon emission is described by the damped-oscillator-type function,[2] $e^{-t/\tau}\{A + B[\cos(\Omega t + \phi)]\}$, where A, B, and ϕ are time-independent constants, τ is the lifetime of the excited states, and Ω is $|E_2 - E_1|/\hbar$, with E_2 and E_1 being the energy of the $M_J = +1$ and -1 states of $^3P_1$, respectively.

A Pierce-type electron gun with an impregnated dispenser-type cathode was constructed in order to obtain an unidirectional electron beam. With the energy near the threshold of excitation, the very low electron current resulted in a very low photon flux. Therefore, the photon-counting technique was employed. The well-distilled Cd metal was sealed into the envelope of the electron gun, which was enclosed in an oven with a quartz window. The electron gun was operated at an oven temperature of about 200°C. A solution of nickel sulfate (14 g) and cobalt sulfate (10 g) in 100 cc of water in a filter (Corning glass type 7-54) filtered the $\lambda = 3261$ Å resonance luminescence corresponding to the transition 7^3P_1 to 7^1S_0. In this filter combination, a bandpass width of 200 Å centered at 3200 Å was quite effective in blocking off the light radiating from the cathode. The electron pulse whose width is about 5 nsec was obtained by pulsing the grid of the electron gun. The $\lambda = 3261$ Å photons were detected through filters and Glan-Thompson quartz polarizer by a photomultiplier tube (RCA type 1P28). To observe the quantum beat phenomena, we used a multichannel delayed-coincidence analyzer to measure the arrival time of photons following the pulse applied to the grid of the electron gun;[3] this gun was pulsed at the rate of 10 kc/sec. Because the count rate was very low, the probability of two photons arriving during the duty cycle of the time-to-pulse height converter was negligible. The schematic of the apparatus is shown in Fig. 1.

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*Published in Phys. Rev. Letters 14, No. 22, 891 (1965).

Figure 2 shows the effect of quantum beats at 0.88 G. The data accumulation time, because of the low count rate, was 10 h.

B. Experiment on 6^3P_2 Hg Metastable State by Modulated-Electron-Beam Excitation

In this experiment, we observed intensity beats in a resonance absorption by coherently excited, nondegenerate, magnetic sublevels of mercury (metastable state 6^3P_2) by modulated electron beam. In order for us to observe these beats, we aimed the modulated electron beam perpendicular to the direction of an externally applied field. For an electron energy near the threshold of excitation of the $^3P_2$ state—if one neglects the electron-exchange effect, and if the modulation is of the type $\mu_0(1 + \epsilon \cos \omega t)$—the intensity of the radiation from the absorbed resonance is proportional to

$$\mu_0 \left[\frac{A}{\gamma} + 2|B| \frac{\gamma \cos X + \omega_{20} \sin X}{\gamma^2 + \omega_{20}^2} \right]$$

$$+ \mu_0 \epsilon |B| \left[\frac{1}{2} \frac{\gamma \cos(\omega t + X) + (\omega - \omega_{20}) \sin(\omega t + X)}{\gamma^2 + (\omega - \omega_{20})^2} \right.$$

$$+ \frac{1}{2} \frac{\gamma \cos(\omega t + X) + (\omega + \omega_{20}) \sin(\omega t + X)}{\gamma^2 + (\omega + \omega_{20})^2}$$

$$\left. + \frac{\gamma \cos(\omega t + X) + \omega \sin(\omega t + X)}{\gamma^2 + \omega^2} \right], \qquad (1)$$

where μ_0, $|A|$, $|B|$, $X(= \arg B)$, and γ (the reciprocal of the spin lifetime τ) are time-independent constants. The first term corresponds to the Hanle depolarization in a magnetic field. The second term represents the beat between the states with $M_J = \pm 2$ and $M_J = 0$ with frequency ω_{20} when the monitoring light is linearly polarized as it propagates along the direction of the field. Equation (1) is derived in a manner similar to that used by Aleksandrov and Kozlov.[4]

In order to observe the beat effect, we had to produce enough of the metastable state. This was accomplished by means of a space-charge-neutralization method in a diode-structure electron gun. By modulating the applied voltage between the cathode and anode, we

could modulate the electron beam at frequencies as high as 30 MHz, although we worked at about 3 MHz with the applied voltage < 20 V.

In this experiment we used the same diode-structure electron gun as we used in our previous neon metastable-state experiment. Because of an objectionable stray magnetic field produced by the heater current for heating the cathode indirectly, we switched the heater current on and off and made observations only when no current was passing through the heater wire. To estimate the bandwidth associated with the beat experiment, we first measured the coherence time, using Hanle effect. The electron beam was modulated at 2.89 MHz, and the absorbed light was detected by a 2.89-MHz tuned amplifier. In order to use a commercially available, lock-in amplifier, we modulated the magnetic field at about 350 Hz. This 350-Hz modulation in the light was first detected by demodulation of 2.89 MHz by a simple diode detector, then by a preamplifier, and finally by the lock-in amplifier. The output of the lock-in amplifier was fed into the digital integrator which, in effect, enabled us to integrate for long periods. The schematic of the experiment is shown in Fig. 3.

Figure 4 shows the 2.89-MHz component of the optical absorption as a function of field corresponding to $m = \pm 2$ to $m = 0$ beat. We also observed a similar signal at twice the field value corresponding to the beat at the Zeeman frequency; the magnitude of this signal, probably arising from the electron exchange, was about half that of the $\Delta m = 2$ beat. The Hanle-effect signal has the same shape but is about 100 times as big. To make certain that the signals were real, we shifted the rf frequency to 2.72 MHz and observed the proper shift of the beat signal as a function of the field.

The magnetic-field bandwidths of the beat signal and Hanle-effect signal were approximately the same. This similarity indicates the same coherence time for the two effects. We observed considerable broadening of the bandwidth as we increased the degree of ionization, indicating a decreased coherence time that was probably due to plasma fields.

References

1. O. Nedelec, M.-N. Deschizeaux, and J.-C. Pebay-Peyroula, Compt. Rend. 257, 3130 (1963).

2. E. B. Aleksandrov, Opt. i Spektroskopiya 16, 377 (1964) [English translation: Opt. Spectry. (USSR) 16, 209 (1964)].

3. W. R. Bennett, Jr., in Advances in Quantum Electronics, edited by J. Singer (Columbia University Press, New York, 1961).

4. E. B. Aleksandrov and Kuzolov, Opt. Spectry. [English translation: Opt. Spectry. (USSR) 16, 289, 580 (1964)].

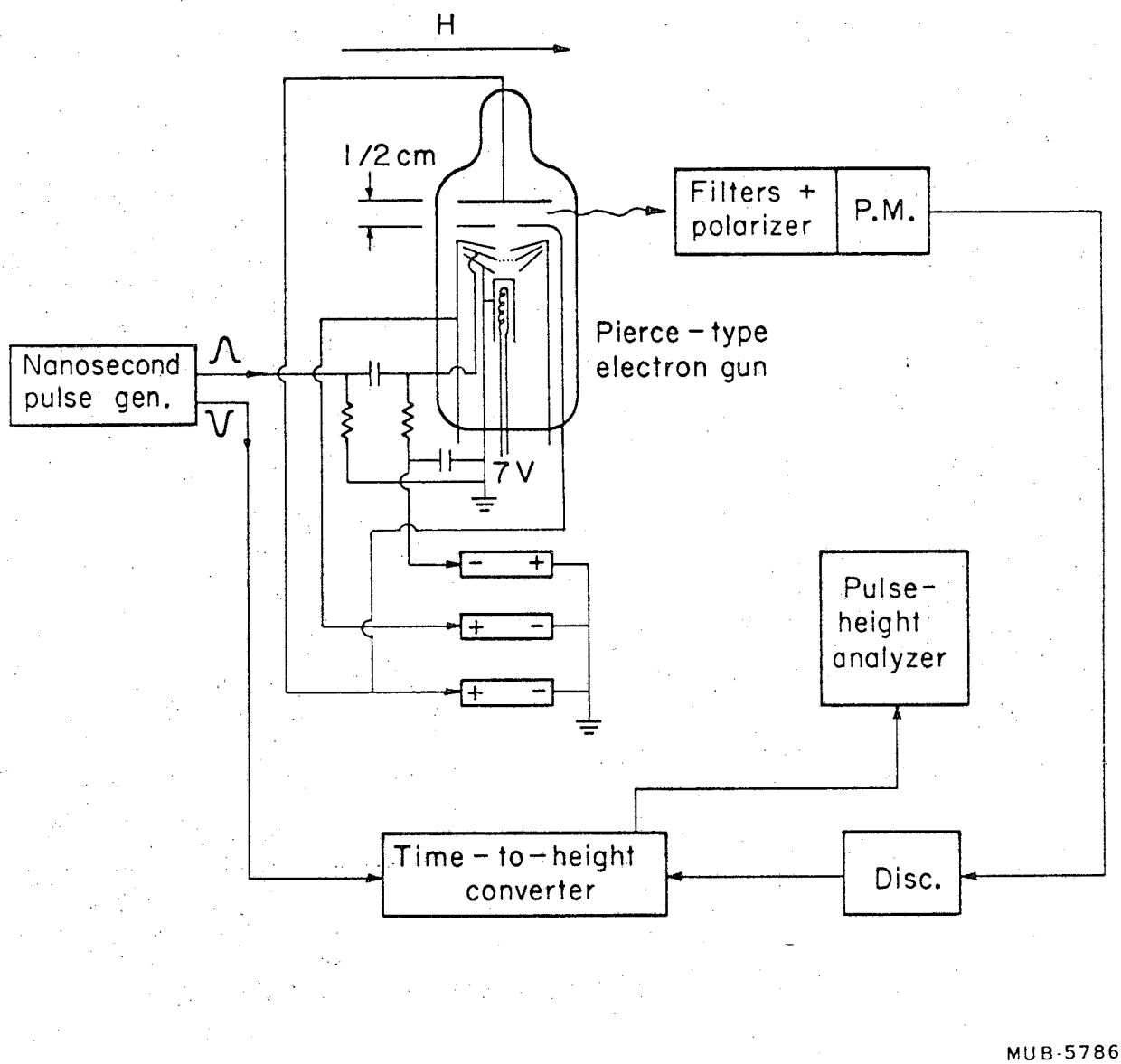
H
1/2 cm
Filters + polarizer
P.M.
Pierce-type electron gun
Nanosecond pulse gen.
7 V
Pulse-height analyzer
Time-to-height converter
Disc.
MUB-5786

Fig. 1

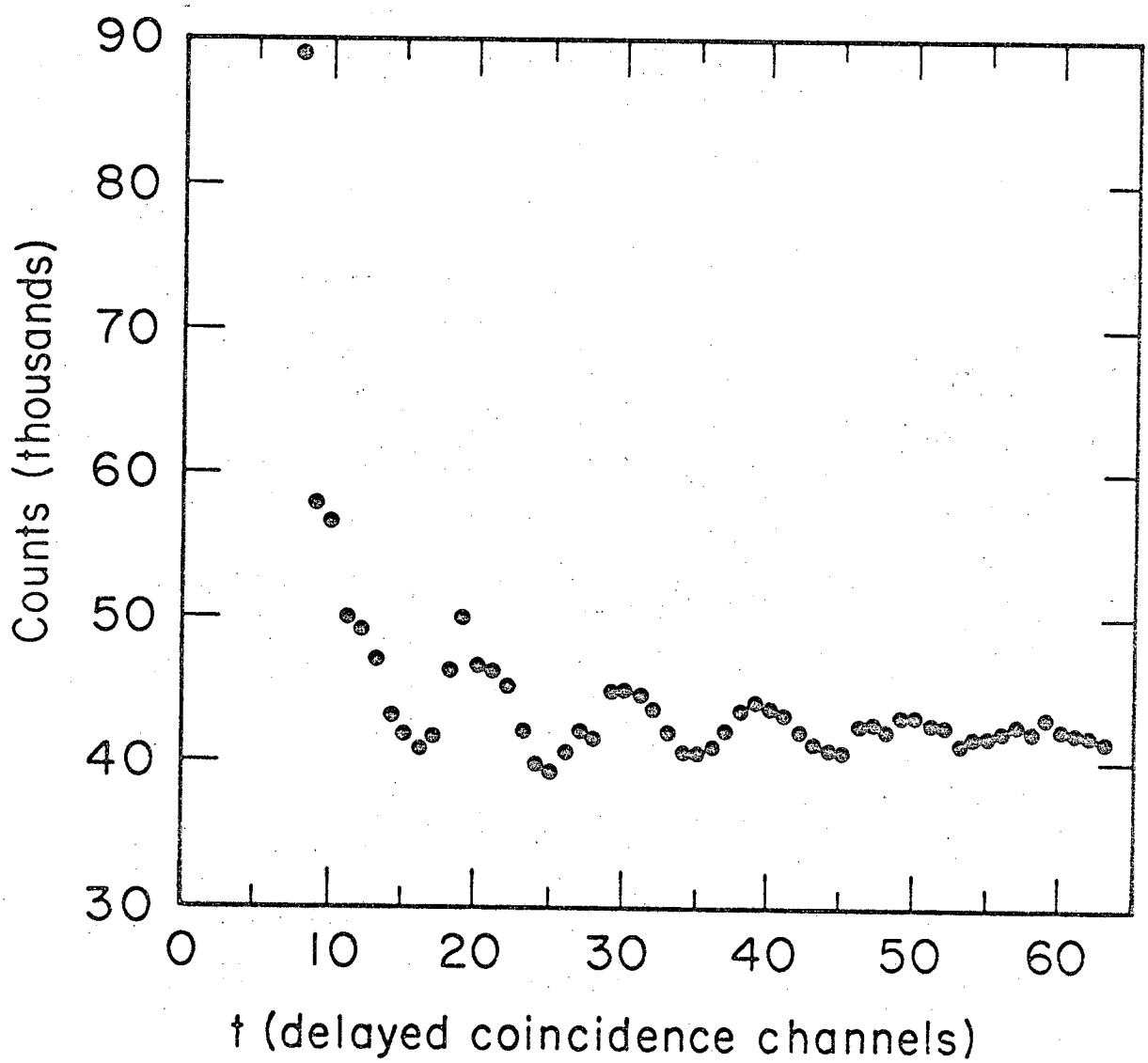

Fig. 2

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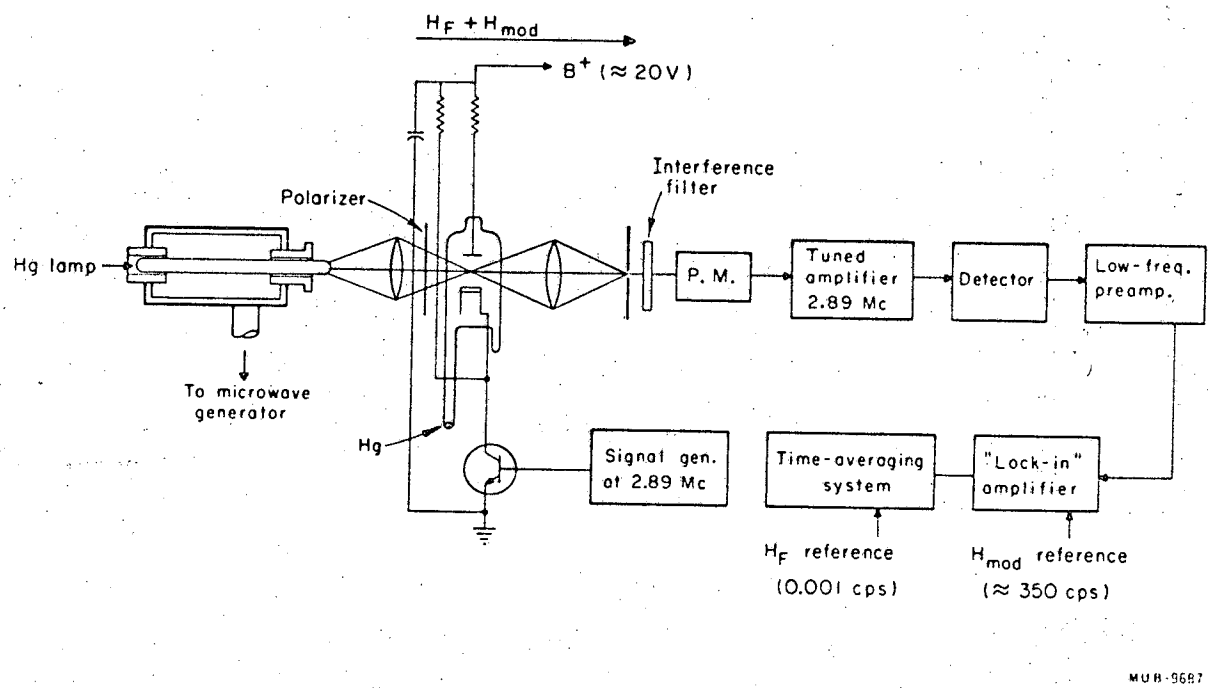
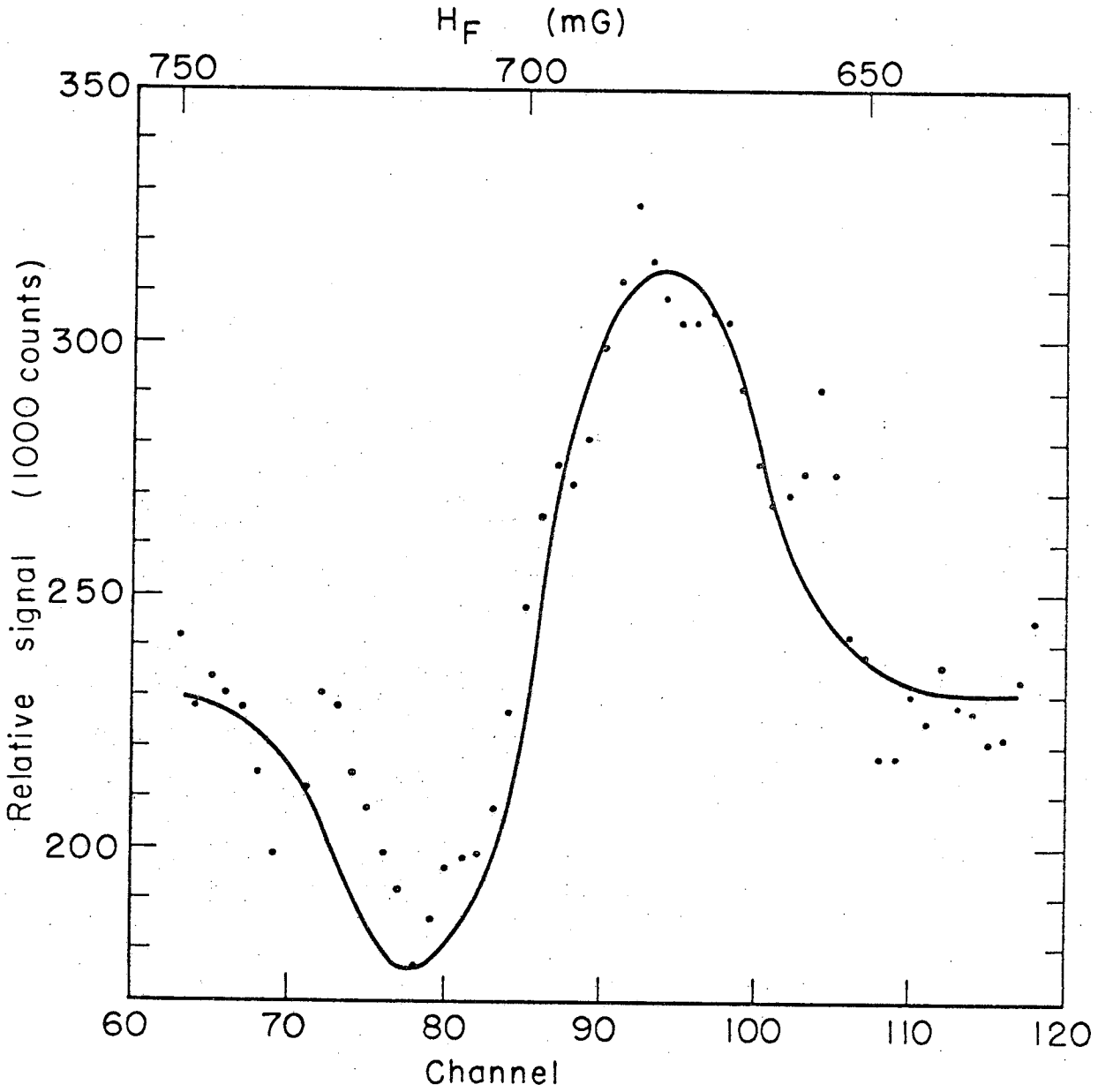

$H_F + H_{mod}$
B^+ (≈ 20V)
Interference filter
Polarizer
Hg lamp
P. M.
Tuned amplifier 2.89 Mc
Detector
Low-freq. preamp.
To microwave generator
Hg
Signal gen. at 2.89 Mc
Time-averaging system
"Lock-in" amplifier
H_F reference (0.001 cps)
H_{mod} reference (≈ 350 cps)
MUB-9687

Fig. 3



MUB-9707

Fig. 4

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