

## Phenomenon of the Metastable State Coherently Excited by Electron Impact\*

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Detailed information is given on an experiment which we have reported previously, studying intensity beats in the resonance absorption due to nondegenerate magnetic sublevels of the  $^3P_2$  mercury metastable state coherently excited by electron impact. Rather straightforward theory is used to explain the experimental result. It is found that regardless of the exciting energy, as long as there is sufficient selective excitation of certain magnetic sublevels, the absorption beat frequency is modulated at twice the Larmor frequency when the incident electron beam is perpendicular to the externally applied field  $H_0$  and the linearly polarized monitoring light is perpendicular to the electron beam.

### I. INTRODUCTION

**I**N this report, we give detailed information on the intensity beats in the resonance absorption due to metastable-state mercury coherently excited by an intense, high-frequency-modulated, unidirectional, low-energy electron beam. This method uses space-charge-neutralized electron flow to our advantage.<sup>1</sup> Similar phenomena have also been observed independently by Nedelec, using a somewhat different method of electron-impact excitation.<sup>2</sup>

Breit has shown the possibility of coherent optical excitation of nondegenerate levels if the exciting resonance radiation is pulsed in a much shorter time than the decay time of the excited state, since under this condition the exciting light has a very broad range of spectral frequency.<sup>3</sup> Thus, provided it is allowed by the optical selection rule, certain magnetic sublevels of the excited states are excited by one wave train. Such excited states exhibit the phenomenon of coherence.

Similar excitation is also possible by electron impact. The broad-band frequency in this case is due to the energy spread of the incident electron. In general, the energy spread of an electron beam is much larger than the energy separations of the nondegenerate levels, so that the necessary condition for coherent excitation is satisfied.

Electron-impact excitation is more useful than optical excitation since *any state*, at least in principle, can be excited by electron impact. For example, the metastable state cannot be optically excited, but it can be excited by electron impact. Therefore, we have performed an experiment on metastable-state Hg to find out if the effect can actually be observed experimentally. The possibility of coherent excitation of the optically excitable state  $^3P_1$  of Cd by electron-impact excitation from  $^1S_0$  to  $^3P_1$  was already experimentally established through the use of a modulated electron beam by Nedelec *et al.*,<sup>2</sup> Aleksandrov,<sup>4</sup> and by our own use of

sharply pulsed electron impact.<sup>5</sup> Therefore, it is obvious that the metastable state should also be coherently excited by electron impact. Nevertheless, we felt such a phenomenon should be observed experimentally. Since we were working with the metastable state, we used optical absorption to detect coherent excitation.

### II. PRINCIPLE OF THE EXPERIMENT

Previous experiments<sup>6</sup> on paramagnetic resonance of the  $^3P_2$  state of Hg have shown that metastable-state atoms excited by electron impact are "aligned." In these experiments, the externally applied magnetic field was parallel to the electron-beam axis. These studies established that the magnetic sublevels with values of  $m_J=0$  and  $\pm 1$  are more selectively excited than  $m_J=\pm 2$  sublevels. However, because of the *cylindrical* symmetry associated about the electron-beam axis, since  $H_0$  is parallel to the beam, these magnetic sublevels are *incoherently* excited; that is, all the nondiagonal density matrix elements vanish. Therefore, the system is describable in terms of diagonal density matrices; that is, in terms of average populations of magnetic sublevels.

If the externally applied field is perpendicular to the electron-beam axis, however, the system of metastable atoms is no longer cylindrically symmetrical around the field. Therefore, the system commences a Larmor precession about the magnetic field. If one monitors such a system with linearly polarized resonance radiation connecting the metastable state, one finds the amount of light absorption is dependent on the relative angle between the linear polarization and the angle of the rotation of the system of atoms due to Larmor precession.

Figure 1 is a pictorial view of the behavior (shown by arrows) of the virtual electric dipole oscillator. Suppose that, at  $t=0$ , the dipole oscillator is created by electron-impact excitation from the  $^1S_0$  ground state to the  $^3P_2$  metastable state parallel to the  $x$  axis. Because of Larmor precession of the atom due to a magnetic dipole

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<sup>1</sup> T. Hadeishi and C.-H. Liu, Phys. Rev. Letters **16**, 1035 (1966).

<sup>2</sup> O. Nedelec, thesis, University of Grenoble, France, 1966 (unpublished).

<sup>3</sup> G. Breit, Rev. Mod. Phys. **5**, 91 (1933).

<sup>4</sup> E. B. Aleksandrov, Opt. i Spektroskopiya **16**, 377 (1964) [English transl.: Opt. Spectry. (USSR) **16**, 209 (1964)].

<sup>5</sup> T. Hadeishi and W. A. Nierenberg, Phys. Rev. Letters **14**, 891 (1965).

<sup>6</sup> H. G. Dehmelt, Phys. Rev. **103**, 1125 (1956); T. Hadeishi, O. McHarris, and W. A. Nierenberg, *ibid.* **138**, A983 (1965).

interaction with  $H_0$ , the electric dipole oscillator precesses about the field. Now suppose that if the light linearly polarized parallel to  $x$  travels along  $H_0$ , the amount of the light absorbed by the metastable state reaches a maximum at  $t=0$ , a minimum at  $t=1/4\omega_0$ , and a maximum again at  $t=1/2\omega_0$ . Since the period of Larmor precession is  $1/\omega_0$ , there are two absorption maxima and two absorption minima per Larmor period. Thus the light absorption is modulated at twice the Larmor frequency while its amplitude is attenuated by  $e^{-\Gamma t}$  due to the finite spin-relaxation time  $\tau$  given by the relation  $\Gamma=1/\tau$ . From this classical point of view, it is rather obvious if a sharply pulsed electron beam is injected at  $t=0, T/2, T, 3T/2, \dots$ , that is, repeated at every half-Larmor period, many metastable-state atoms would precess about the field synchronously (coherently). Because of the large concentration of metastable-state atoms exhibiting coherent behavior, the phenomenon is observable by absorption of resonance radiation.

Although a very elegant theory of light beats has been formulated by Nedelec,<sup>2</sup> we shall present a more straightforward theory to describe the phenomena observed, emphasizing intuition without involving a great deal of complex mathematical theory.

### III. THEORY

Although previous experiments<sup>6</sup> on paramagnetic resonance of the  $^3P_2$  state of Hg definitely showed that the states with  $|m\rangle=|0\rangle$  and  $|\pm 1\rangle$  are more selectively produced than  $|m\rangle=|\pm 2\rangle$ , there is no way of knowing by just what amount these states are produced by the electron-impact method used in these experiments. All we know is that if we take the electron-beam axis as an axis of quantization, the  $^3P_2$  state's wave function  $\Psi$  can be represented as

$$\Psi = a_2(t)|^3P_{2,2}\rangle + a_1(t)|^3P_{2,1}\rangle + a_0(t)|^3P_{2,0}\rangle + a_{-1}(t)|^3P_{2,-1}\rangle + a_{-2}(t)|^3P_{2,-2}\rangle, \quad (1)$$

such that  $\rho_{22}=\rho_{-2,-2}$ ,  $\rho_{11}=\rho_{-1,-1}$ , and  $\rho_{00}$  and  $\rho_{11}$  are greater than  $\rho_{22}$ , where  $\rho_{mm}$  is the diagonal density matrix element defined as  $\rho_{mm}=\langle a_m^*(t)a_m(t)\rangle$ . Also, in this coordinate system,  $a_2(t)$ ,  $a_1(t)$ ,  $a_0(t)$ ,  $a_{-1}(t)$ , and  $a_{-2}(t)$  are completely independent, so that all the nondiagonal density matrices vanish. However, because of the observed fact that  $\rho_{00}, \rho_{11} > \rho_{22}$ , we are certainly justified in assigning the production rate  $\alpha$ ,  $\beta$ , and  $\gamma$  to indicate the rate of production of the state with  $m=\pm 2$ ,  $m=\pm 1$  and 0. Therefore, the initial magnetic sublevel populations at the instant of excitation can be represented by

$$(\rho_0) = \begin{pmatrix} \alpha & 0 & 0 & 0 & 0 \\ 0 & \beta & 0 & 0 & 0 \\ 0 & 0 & \gamma & 0 & 0 \\ 0 & 0 & 0 & \beta & 0 \\ 0 & 0 & 0 & 0 & \alpha \end{pmatrix} \frac{1}{(2\alpha+2\beta+\gamma)}. \quad (2)$$

In addition, it was found experimentally that the selective excitation rate is independent of the strength of the externally applied magnetic field  $H_0$ . Suppose the electron beam is injected perpendicular to  $H_0$ ; if we take the direction of  $H_0$  as an axis of quantization,  $\rho_0$  in this new coordinate system transforms to  $\rho^0$  by the similarity transformation given by

$$\rho^0 = \mathfrak{D}^{(2)}(0, \pi/2, 0)\rho_0\mathfrak{D}^{(2)\dagger}(0, \pi/2, 0), \quad (3)$$

where  $\mathfrak{D}^{(2)}$  is the angular rotation operator for  $J=2$ . The time-rate change of the density matrix is then given by

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[\mathfrak{H}, \rho] - \Gamma\rho + \rho^0\frac{1}{T_p}, \quad (4)$$

where  $T_p$  is the electron pumping speed (similar to that of an optical pumping speed),  $(-i/\hbar)[\mathfrak{H}, \rho]$  describes the process of time evolution of the excited state due to interaction of the magnetic dipole moment with the field,  $\Gamma\rho$  is the decay rate due to such factors as collisions with other atoms, and  $\rho^0/T_p$  describes the process of the excitation. Since the Hamiltonian  $\mathfrak{H}$  in Eq. (4) is equal to  $-\mathbf{u}\cdot\mathbf{H}_0$ , Eq. (4) can be written as

$$\frac{d\rho_{mm'}}{dt} = -i(m-m')\omega_0\rho_{mm'} - \Gamma\rho_{mm'} + \rho_{mm'}^0\frac{1}{T_p}, \quad (5)$$

where  $\omega_0$  is the Larmor angular frequency.

The time-dependent intensity of the light absorbed was derived by Barrat,<sup>7</sup> and is given by

$$I(t) = A \sum_{mm'\mu} \langle \mu | \mathbf{e}_\lambda \cdot \mathbf{D} | m \rangle \rho_{mm'}(t) \langle m' | \mathbf{e}_\lambda \cdot \mathbf{D} | \mu \rangle, \quad (6)$$

where  $A$  is a constant multiplication factor and  $\mathbf{D}$  is the vector electric dipole operator. Thus  $\mathbf{e}_\lambda \cdot \mathbf{D}$  denotes the polarization of the light in the  $\mathbf{e}_\lambda$  direction.

Let us consider the case in which the incident electron beam is modulated periodically as  $(1+\cos\omega t)$ . Then Eq. (5) can be written as

$$\frac{d\rho_{mm'}}{dt} = \frac{1}{T_p} \rho_{mm'}^0 (1 + \cos\omega t) - i(m-m')\omega_0\rho_{mm'} - \Gamma\rho_{mm'}. \quad (7)$$

The solution of this equation, by noting that

$$(1 + \cos\omega t) = [1 + (e^{i\omega t}/2) + (e^{-i\omega t}/2)],$$

is given by

$$\rho_{mm'}(t) = \frac{1}{T_p} \rho_{mm'}^0 \left[ \frac{1}{\Gamma + i(m-m')\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma + i\omega + i(m-m')\omega_0} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma - i\omega + i(m-m')\omega_0} \right]. \quad (8)$$

Before we substitute Eq. (8) into Eq. (6), it is conven-

<sup>7</sup> J. P. Barrat, J. Phys. Radium 20, 541 (1959); 20, 633 (1959); 20, 657 (1959).

ient at this stage to calculate  $\rho_{mm}^0$  from Eq. (3). The rotation operator  $\mathfrak{D}^{(2)}(0, \pi/2, 0)$  is given as

$$\mathfrak{D}^{(2)}\left(0, \frac{\pi}{2}, 0\right) = \begin{bmatrix} \frac{1}{4} & -2/4 & \frac{1}{4}(\sqrt{6}) & -2/4 & \frac{1}{4} \\ 2/4 & -2/4 & 0 & 2/4 & -2/4 \\ \frac{1}{4}(\sqrt{6}) & 0 & -\frac{1}{2} & 0 & \frac{1}{4}(\sqrt{6}) \\ 2/4 & 2/4 & 0 & -2/4 & -2/4 \\ \frac{1}{4} & 2/4 & \frac{1}{4}(\sqrt{6}) & 2/4 & \frac{1}{4} \end{bmatrix}. \quad (9)$$

From Eq. (3) we get

$$(\mathfrak{D}^{(2)}(0, \pi/2, 0)\rho_0\mathfrak{D}^{(2)\dagger}(0, \pi/2, 0)) = \begin{bmatrix} \frac{1}{8}(\alpha+4\beta+3\gamma) & 0 & \frac{1}{8}(\sqrt{6})(\alpha-\gamma) & 0 & \frac{1}{8}(\alpha-4\beta+3\gamma) \\ 0 & \frac{1}{2}(\alpha+\beta) & 0 & \frac{1}{2}(\alpha-\beta) & 0 \\ \frac{1}{8}(\sqrt{6})(\alpha-\gamma) & 0 & \frac{1}{4}(3\alpha+\gamma) & 0 & \frac{1}{8}(\sqrt{6})(\alpha-\gamma) \\ 0 & \frac{1}{2}(\alpha-\beta) & 0 & \frac{1}{2}(\alpha+\beta) & 0 \\ \frac{1}{8}(\alpha-4\beta+3\gamma) & 0 & \frac{1}{8}(\sqrt{6})(\alpha-\gamma) & 0 & \frac{1}{8}(\alpha+4\beta+3\gamma) \end{bmatrix} \frac{1}{2\alpha+2\beta+\gamma}. \quad (10)$$

Substituting Eq. (10) into Eq. (8) we get

$$\begin{aligned} \rho_{22}(t) &= \xi \frac{1}{8}(\alpha+4\beta+3\gamma) \left[ \frac{1}{\Gamma} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i\omega} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i\omega} \right] = \rho_{-2-2}, \\ \rho_{20}(t) &= \xi \frac{1}{8}(\sqrt{6})(\alpha-\gamma) \left[ \frac{1}{\Gamma+i2\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i(\omega+2\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i(\omega-2\omega_0)} \right] = \rho_{0-2}, \\ \rho_{00}(t) &= \xi \frac{1}{4}(3\alpha+\gamma) \left[ \frac{1}{\Gamma} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i\omega} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i\omega} \right], \\ \rho_{11}(t) &= \xi \frac{1}{2}(\alpha+\beta) \left[ \frac{1}{\Gamma} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i\omega} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i\omega} \right] = \rho_{-1-1}, \\ \rho_{1-1}(t) &= \xi \frac{1}{2}(\alpha-\beta) \left[ \frac{1}{\Gamma+i2\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i(\omega+2\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i(\omega-2\omega_0)} \right], \\ \rho_{-20}(t) &= \xi \frac{1}{8}(\sqrt{6})(\alpha-\gamma) \left[ \frac{1}{\Gamma-i2\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i(\omega-2\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i(\omega+2\omega_0)} \right] = \rho_{02}, \\ \rho_{-11}(t) &= \xi \frac{1}{2}(\alpha-\beta) \left[ \frac{1}{\Gamma-i2\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i(\omega-2\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i(\omega+2\omega_0)} \right], \\ \rho_{2-2} &= \xi \frac{1}{8}(\alpha-4\beta+3\gamma) \left[ \frac{1}{\Gamma+i4\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i(\omega+4\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i(\omega-4\omega_0)} \right], \\ \rho_{-2-2} &= \xi \frac{1}{8}(\alpha-4\beta+3\gamma) \left[ \frac{1}{\Gamma-i4\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i(\omega-4\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma-i(\omega+4\omega_0)} \right], \end{aligned} \quad (11)$$

and

where

$$\xi = [1/(2\alpha+2\beta+\gamma)](1/T_p).$$

Next we consider observing the resonance absorption of linearly polarized light polarized parallel to the  $x$  direction and propagating along the externally applied magnetic field. Therefore, the matrix element of the dipole

operator we must calculate is of the type

$$\langle \mu | \frac{rC_{-1}^1 - rC_{+1}^1}{\sqrt{2}} | m \rangle,$$

where  $C_q^1$  is the spherical tensor of rank 1 defined as  $C_{\pm 1}^1 = \mp \sqrt{\frac{1}{2}}(x \pm iy)/r$  and  $C_0^1 = z/r$ . The matrix elements can be most readily calculated with the Wigner-Eckart theorem by noting

$$\langle \mu | C_q^1 | m \rangle = \langle {}^3S_1, J=1, \mu | C_q^1 | {}^3P_2, J=2, m \rangle = (-1)^{1-\mu} \begin{pmatrix} 1 & 1 & 2 \\ -\mu & q & m \end{pmatrix} \langle {}^3S_1, J=1 || C^1 || {}^3P_2, J=2 \rangle, \quad (12)$$

where

$$\begin{pmatrix} 1 & 1 & 2 \\ -\mu & q & m \end{pmatrix}$$

is the  $3j$  symbol and  $\langle {}^3S_1, J=1 || C^1 || {}^3P_2, J=2 \rangle$  is the reduced matrix element.<sup>8</sup> Evaluating the appropriate matrix element of the type in Eq. (12), and substituting Eqs. (11) and (12) into Eq. (6), we get

$$\begin{aligned} I(t) &= A'(57\alpha + 36\beta + 37\gamma) \left[ \frac{1}{\Gamma} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma + i\omega} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma - i\omega} \right] + B'[\sqrt{6(\alpha - \gamma)} + 6(\alpha - \beta)] \\ &\quad \times \left\{ \left[ \frac{1}{\Gamma + i2\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma + i(\omega + 2\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma - i(\omega - 2\omega_0)} \right] + \left[ \frac{1}{\Gamma - i2\omega_0} + \frac{\frac{1}{2}e^{i\omega t}}{\Gamma + i(\omega - 2\omega_0)} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma - i(\omega + 2\omega_0)} \right] \right\} \\ &= A' \left[ \frac{1}{\Gamma} + \frac{2 \cos(\omega t - \phi_0)}{(\Gamma^2 + \omega^2)^{1/2}} \right] (57\alpha + 36\beta + 37\gamma) + B' \left[ \frac{2 \cos \phi_0}{(\Gamma^2 + 2\omega_0^2)^{1/2}} \right] \{ \sqrt{6(\alpha - \gamma)} + 6(\alpha - \beta) \} \\ &\quad + B' \left[ \frac{2 \cos(\omega t - \phi_1)}{[\Gamma^2 + (\omega + 2\omega_0)^2]^{1/2}} + \frac{2 \cos(\omega t - \phi_2)}{[\Gamma^2 + (\omega - 2\omega_0)^2]^{1/2}} \right] \{ \sqrt{6(\alpha - \gamma)} + 6(\alpha - \beta) \}, \quad (13) \end{aligned}$$

where

$\tan \phi_0 = \omega/\Gamma$ ,  $\tan \phi_1 = (\omega + 2\omega_0)/\Gamma$ ,  $\tan \phi_2 = (\omega - 2\omega_0)/\Gamma$ , and  $A'$  and  $B'$  are time-independent constants.

Thus we obtain an expected result. The first term shows the uninteresting case of modulation of the resonance-absorption light with the electron-beam modulation that is always expected to occur.

The second term shows the depolarization effect in the magnetic field, known as the Hanle effect. The third term shows the resonance behavior in the modulation amplitude of the absorbed light when  $\omega = 2\omega_0$  and  $\omega = -2\omega_0$ . The term  $\omega = -2\omega_0$  shows the effect of reversing the direction of the magnetic field.

Equation (13) also satisfies the case in which we expect no observable beat phenomenon if the production rate is  $\alpha = \beta = \gamma$ ; that is, there is no selective excitation of the Zeeman magnetic sublevel. *Therefore, selective excitation must be achieved in order to observe the beat phenomenon.*

#### IV. EXPERIMENTAL CONSIDERATIONS

As Eq. (13) shows, we must first of all achieve selective excitation of the Zeeman sublevels so that

<sup>8</sup> See, for example, B. R. Judd, *Operator Techniques in Atomic Spectroscopy* (McGraw-Hill Book Company, Inc., New York, 1963).

$\alpha - \gamma$  and  $\alpha - \beta$  are as large as possible. From the consideration of inelastic electron-atom collisions,<sup>9</sup> this can be achieved with a low-energy, unidirectional electron beam. In order to satisfy the condition  $\omega = 2\omega_0$  so that  $\omega_0 \gg 1/\tau$  (where  $\tau$  is the metastable-state spin-

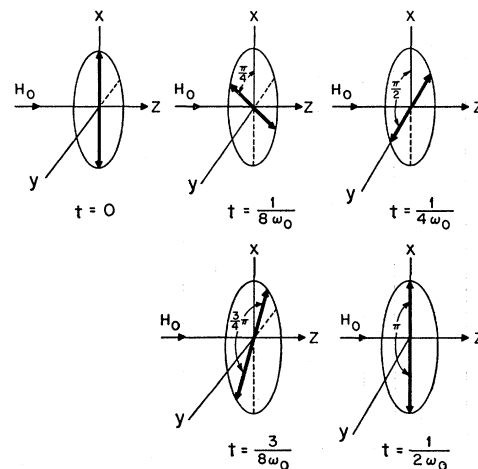


FIG. 1. Pictorial view of the precession of the electric dipole oscillator.

<sup>9</sup> See, for example, I. C. Percival and M. J. Seaton, *Phil. Trans. Roy. Soc. (London)* **251**, 113 (1958); L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), pp. 205-209; W. E. Lamb, *Phys. Rev.* **105**, 559 (1957).

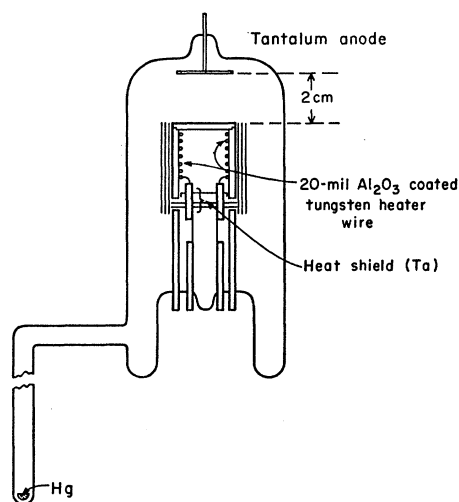


FIG. 2. Cross-sectional view of the excitation tube.

relaxation time of the order of  $10^{-4}$  sec that we initially measured), the electron beam should be modulated at a few MHz. Furthermore, to observe the resonance absorption, we must have a high concentration of the metastable state.

Therefore, the electron beam should have the following properties:

(a) The electron beam must be unidirectional; the vapor pressure of the mercury must then be not more than  $1 \mu$  to avoid the multiple scattering that alters the directionality of the electron beam.

(b) To produce selective excitation, the energy of the electron beam must be low.

(c) To produce a high concentration of metastable-state atoms, electron-beam current must be high.

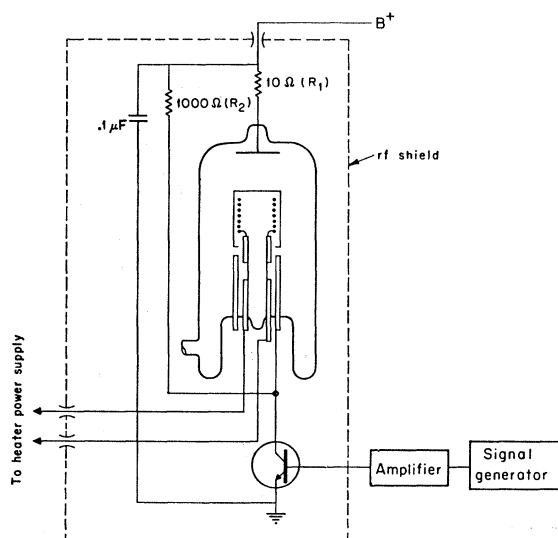
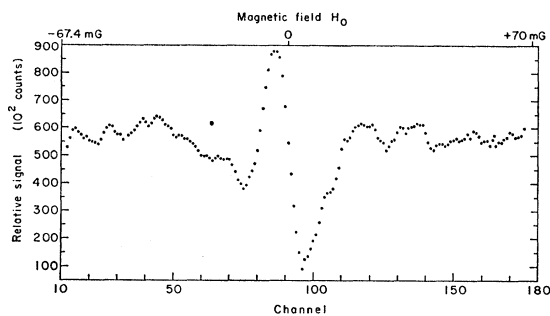
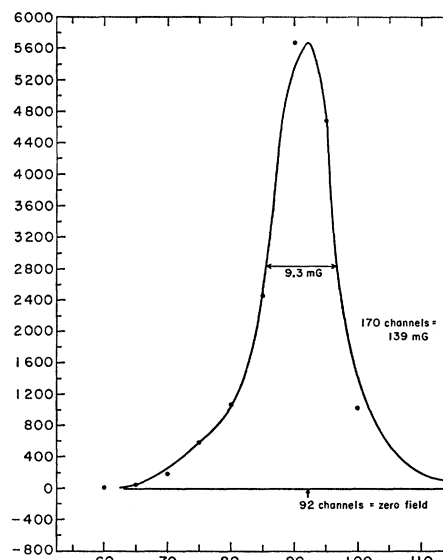


FIG. 3. Method used to modulate the space-charge-neutralized electron beam at high frequency ( $\approx 3$  MHz).



(a)



(b)

FIG. 4. Hanle-effect signal. (a) Experimentally observed signal (b) Integrated curve of (a).

(d) The electron beam must be modulated at high frequency.

Fortunately, the first three requirements can be easily met by the space-charge-neutralized electron flow that was theoretically and experimentally shown by the brilliant work of Langmuir.<sup>10</sup> In order to achieve high-frequency modulation of a space-charge-neutralized, high-current electron beam, we interrupted the electron flow from the cathode to the ground, using as a switch a fast-response transistor capable of drawing a high current. We found we were able to modulate the electron beam 100% with a peak-to-peak current of 400 mA at 3 MHz.

A simple diode-structure electron-gun excitation tube, saturated with Hg vapor at room temperature, was used for this experiment. In order to avoid cathode-surface damage by back-bombardment of  $\text{Hg}^+$ , we used a Phillips cathode that has a diameter of  $\frac{3}{4}$  in., and was indirectly heated.

<sup>10</sup> I. Langmuir, Phys. Rev. 33, 954 (1929).

There are several reasons for using this method. First of all, we achieve space-charge-neutralized electron flow because of charged "double sheaths" near the cathode surface—the  $\text{Hg}^+$  sheath and the electron sheath.<sup>10</sup> The recombination time of  $\text{Hg}^+$  is a few msec; Therefore, when we switched the tube at the 3-MHz rate, the recombination time is almost infinite compared with the switching period. By means of  $R_2$ , the cathode potential becomes equal to the anode potential when the switch is open; thus we expect a very small amount of current to flow from cathode to anode. On the other hand, because of the long  $\text{Hg}^+$  recombination time, the double sheath quickly returns to dc behavior when the switch is closed, so that a large space-charge-neutralized electron current flows to the anode. Since the resistance across the tube is much less than that of  $R_2$  when the switch is closed, most of the current flows through the diode rather than through  $R_2$ . The capacitor  $C$  is used to short out the transient effect when the switch is suddenly opened.

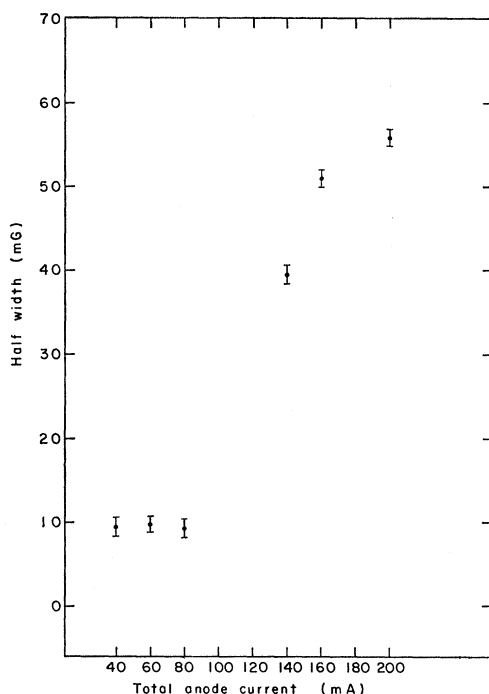


FIG. 5. Dependence of the spin-relaxation time on the electron-beam current at room temperature. The relative sign of the signals is reversed for current higher than 140 mA.

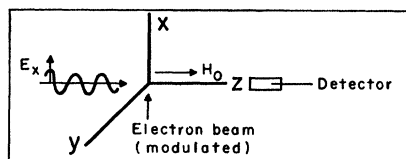


FIG. 6. Relative directions of the resonance radiation, electron beam and the applied field.

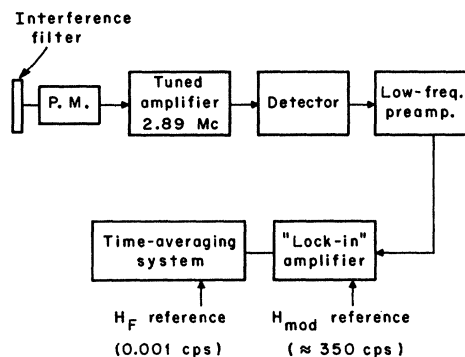


FIG. 7. Block diagram of detection system.

To eliminate the stray magnetic field produced by the indirect cathode heater, observations were made only when the heater current was turned off. Because of the massive cathode we used, we could turn the heater current on and off every 1/50 sec with only a negligible drop in the electron current while the heater current was off.

Although the lifetime of the metastable state of  $\text{Hg}$  is typically a few msec, the spin-relaxation time is much shorter. Spin-relaxation time can be readily measured either by the bandwidth associated with paramagnetic resonance of the metastable state or by the magnetic bandwidth associated with the Hanle effect.

Since the experimental setup for observation of the Hanle effect satisfies experimental conditions for the beat experiment, we first observed the Hanle effect to determine the spin-relaxation time. The main reason for performing the Hanle-effect experiment was to determine a reasonable modulation frequency, since  $\omega \gg 1/\tau$  where  $\tau$  is the spin-relaxation time. Figure 2 shows the cross-sectional view of the diode structure electron gun. Figure 3 shows the method we used to

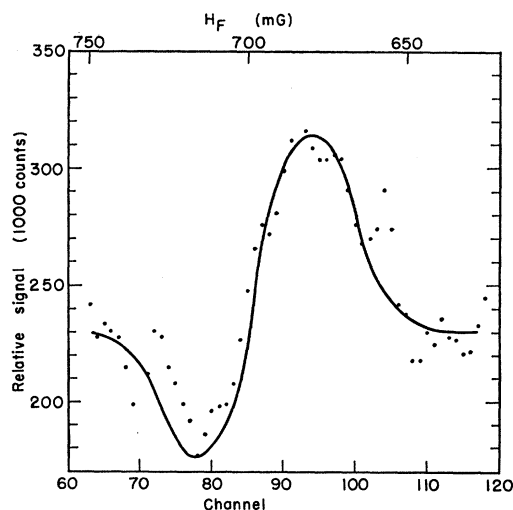


FIG. 8. Resonance-absorption beat signal with electron beam modulation at 2.89 MHz.

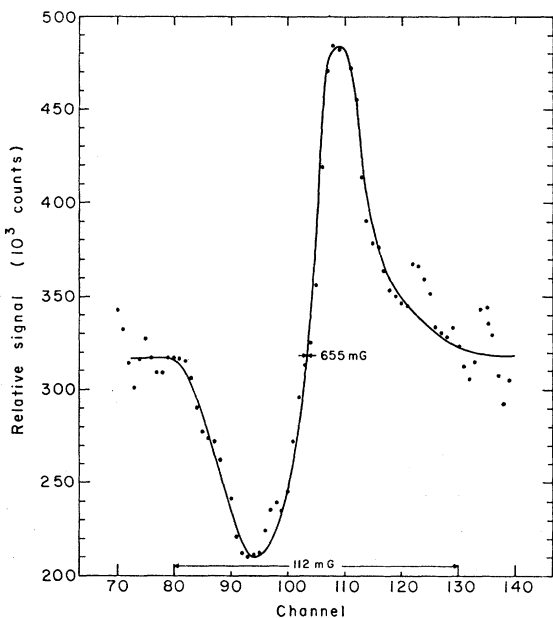


Fig. 9. Resonance-absorption beat signal with electron beam modulation at 2.72 MHz.

modulate the space-charge-neutralized electron beam at high frequency. Figure 4 show a typical Hanle-effect signal. The spin-relaxation time, determined from the curve at  $J=31$  mA/cm<sup>2</sup> at room temperature is,  $2 \times 10^{-4}$  sec. This is done by matching the curve of Fig. 4(b) to the second term of Eq. (13). The reason for the dispersion-type curve in Fig. 4 is that magnetic-field modulation is used in order to employ a phase-sensitive detector that eliminates the 50 Hz "chopping" frequency synchronized with the heater "chopping" frequency. Thus, signal is fed into the detection system only while no current flows through the indirect cathode-heater wire. Figure 5 shows the dependence of the spin-relaxation time on the electron current.

Since  $\tau \approx 10^{-4}$  sec, we decided to use 2.89 MHz for electron-current modulation since we had a narrow-band amplifier tuned to this frequency from another experiment. Since it is more convenient to keep the frequency fixed, we swept the magnetic field while keeping the modulation frequency fixed.

The modulated electron beam is injected perpendicular to the externally applied magnetic field  $H_0$ ; the linearly polarized monitoring resonance radiation  $\lambda = 5461 \text{ \AA}$  ( $6^3P_2-7^3S_1$ ), propagating along the magnetic field  $H_0$ , is used to detect the behavior of coherently excited metastable-state atoms (Fig. 6). Figure 7 shows the detection system described previously.<sup>1</sup> Figures 8 and 9 show the high-frequency component of the optical absorption as a function of applied magnetic field  $H_0$ . These two curves correspond to  $\omega = 2\omega_0$ .

## V. CONCLUSION

Since the applied magnetic field was fairly small, no correction for the curvature of the electron beam was considered. By matching the Hanle-effect signal to the term corresponding to the Hanle-effect term in Eq. (13), we could measure the spin-relaxation time quite readily. This may perhaps be a convenient way of measuring the relaxation time of the nonradiative metastable state as a function of environmental conditions such as pressure, and degree of ionization.

However, the main purpose of this experiment was to observe the beat phenomenon. The phenomenon we observed is, in principle, quite similar to that observed by Bell and Bloom<sup>11</sup> using an optical pumping method. However, the optical method works only with states connected by the electric dipole transition, while the electron-impact method removes this fundamental restriction associated with the optical selection rule. An additional advantage in using space-charge-neutralized, high-flux electron flow is that "electron pumping speed" is much faster than "optical pumping speed" since by the former method electron current can be increased up to 10 A/cm<sup>2</sup>.

Perhaps the most important feature of this method is that radiofrequency fields, which perturb the excited state, are not necessary for obtaining information on that state. By modulating the electron beam at a hyperfine-structure frequency, this method should also yield information on the hyperfine structure of isotopic atoms.

<sup>11</sup> W. Bell and A. Bloom, Phys. Rev. Letters **6**, 280 (1961).