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Berkeley, California

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Tetsuo Hadeishi

March 13, 1967

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Tetsuo Hadeishi

Lawrence Radiation Laboratory
University of California
Berkeley, California

Abstract

We observed intensity beats in the resonance luminescence arising from the interference of electromagnetic-field quanta that are emitted from the coherently excited nondegenerate magnetic sublevels of the excited state of atoms that are formed by sharply pulsed, low-energy, unidirectional electron beams.[1] In a metastable state, a similar effect was obtained by monitoring the beats in the resonance absorption.[2] Since a large concentration of the coherently excited metastable states are required in the absorption experiment, a space-charge neutralized, high-flux unidirectional, low-energy, high-frequency modulated electron beam was used. We performed these experiments (1) to find out if the phenomena are observable and (2) to offer new method of investigation of excited atoms.

Introduction

Breit[3] has shown a possibility of coherent excitation of nondegenerate levels if the exciting resonance radiation is pulsed in a time much shorter than the decay time of the excited states. A reason for this is that the shorter the light pulse, the broader the range of the spectral frequency. Therefore, if the width of the line $\Delta\nu$ is large compared with the natural width and the separation of the nondegenerate levels, these levels are excited in a time of order $\Delta t \approx \frac{1}{\Delta\nu} << \tau$, where τ is the natural lifetime of the excited state. This means all the nondegenerate states are excited by one wave packet, provided the excitations are allowed by the optical selection rule. Under such excitation conditions, the phase differences among the nondegenerate levels are perfectly definite. Such excited states are called coherently excited states. The interference of resonance radiations emitted from coherently produced states results in beat phenomena.

A similar condition can be satisfied by the electron-impact excitation. In general, the electron-beam energy is not monoenergetic. For nondegenerate levels that are Zeeman magnetic sublevels, the necessary condition that the spread of the electron energy ΔE be larger than the Zeeman sublevel separation under the weak externally applied magnetic field is well satisfied. In addition to this requirement, there must be preferential or selective excitation of the certain Zeeman sublevels in order to induce coherence among the Zeeman sublevels. Unlike the case of optical excitation, the selection rule associated with the electron beam is less definite. However, the selective excitation of Zeeman sublevels by electron impact depends on the direction of the incident electron and its momentum. Since we can control the momentum of the electron quite readily by merely changing the acceleration voltage, we can satisfy essentially the same condition as that of excitation by light, with one definite advantage—any state can be excited by electron impact.

By electron-impact excitation, the excited atoms are "aligned," i.e., the sublevels with M and -M are produced with equal probability. In order to illustrate how the experiment works, let us consider $^1S_0$-$^3P_1$ electron-impact excitation of Cd atoms from the ground state to the first excited state. The $^3P_1$ state decays to the $^1S_0$ ground state by emitting λ at 3261 Å photons. Suppose that the energy of the incident electron is only slightly higher than the threshold energy of $^3P_1$ state, and the angular momentum of the incident electron is zero before the collision; then the angular momentum of the scattered

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electron is zero, since the electron's linear momentum is zero after the collision. From considering the conservation of angular momentum and neglecting the effect of electron exchange, we expect the state with $m = 0$ to be selectively excited. If the externally applied magnetic field is parallel to the electron beam direction, we expect no change in the selective excitation of the $m = 0$ state. However, suppose the magnetic field is applied perpendicular to the beam direction; by taking this direction as the axis of quantization, we find that the state that was originally in $m = 0$ transforms to a linear combination of $m = +1$ and $m = -1$ states. Their relative phase must be perfectly definite since in the original coordinate (the one in which the quantization axis is taken as the electron beam direction), m must be equal to zero. Now in the new coordinate system, the excited state with $m = 1$ and $m = -1$ coherently produced commences its Larmor precession about the magnetic field while maintaining its relative phase between $m = 1$ and $m = -1$ in the absence of other perturbations such as collision with other atoms. Now if one would observe the light radiated from the $^3P_1$ state due to the spontaneous transition to the $^1S_0$ ground state (say in the direction along the magnetic field), he would see that the linearly polarized light would rotate with the Larmor frequency. So if one would observe the transmission of the light through the linear polarizer, he would see that the intensity of the light is modulated at twice the Larmor frequency.

In the metastable state, one can also observe a similar effect in terms of absorption of linearly polarized resonance radiation passing through the assembly of atoms that are coherently excited by the electron impact. However, in a metastable state experiment, a large concentration of metastable atoms is required since a detectable amount of absorption of resonance radiation is necessary.

Theory

We shall consider only a rather idealized case since more general considerations take rather lengthy derivation. The wave function (ψ) of the excited state immediately after electron impact is given by

$$\psi(t=0) = \sum_m a_m(t=0)\,|m\rangle. \tag{1}$$

If we take the electron beam axis as the quantization axis, the phases of a_m are completely random because of the cylindrical symmetry about the beam axis. Therefore $\rho_{0mm'}(t=0) = \overline{a_m^*(t=0)a_{m'}^*(t=0)} = 0$ if $m \neq m'$ (where $\rho_{0mm'}$ is the density matrix). However, if one would apply magnetic field at some angle with respect to the electron beam, the density matrix ρ^o in this new coordinate would be given by transformation of the coordinate

$$\rho^o = \rho(t=0) = R\,\rho_o R^\dagger. \tag{2}$$

where R is the rotation operator of the angular momentum. Therefore, in this coordinate $\rho^o_{mm'} \neq 0$ for certain m and m'.

The intensity of light that is emitted from the excited state is given[4] by

$$I(t) = I_o \Gamma \sum_{mm'\mu} \langle \mu | \hat{e}_\lambda^* \cdot \vec{D}^\dagger | m\rangle\, \rho_{mm'}(t) \langle m' | \hat{e}_\lambda \cdot \vec{D} | \mu\rangle, \tag{3}$$

where $\vec{D}^\dagger$ is the dipole operator, $|\mu\rangle$ the ground-state Zeeman level, $|m\rangle$ the excited-state Zeeman level and $\Gamma = \frac{1}{\tau}$, where τ is the lifetime of the excited state.

When light is absorbed, the expression is quite similar with the exception of a constant multiplying factor. This is given by[5]

$$I(t) = \frac{k_o}{\hbar}\, \rho_s(k_o) \sum_{mm'\mu} \langle \mu | \vec{e}_\lambda^* \cdot \vec{D}^\dagger | m\rangle\, \rho_{mm'}(t) \langle m' | \vec{e}_\lambda \cdot \vec{D} | \mu\rangle. \tag{4}$$

Intensity Beats in Resonance Fluorescence

We shall consider an electron-impact excitation of a Cd atom from the $^1S_0$ ground state to the $^3P_1$ first excited state that decays to the ground state with emission of $\lambda = 3261$ Å resonance radiation. When the electron beam is at the threshold energy of excitation and the state with $|m\rangle = |0\rangle$ is assumed to be the only state that is produced, then in the new coordinate in which the external magnetic field is applied perpendicularly to the electron beam, we obtain from Eq. (2)

$$\rho^o = \begin{pmatrix} \frac{1}{2} & -\frac{1}{\sqrt{2}} & \frac{1}{2} \\ \frac{1}{\sqrt{2}} & 0 & -\frac{1}{\sqrt{2}} \\ \frac{1}{2} & \frac{1}{\sqrt{2}} & \frac{1}{2} \end{pmatrix} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \frac{1}{2} & \frac{1}{\sqrt{2}} & \frac{1}{2} \\ -\frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ \frac{1}{2} & -\frac{1}{\sqrt{2}} & \frac{1}{2} \end{pmatrix} = \begin{pmatrix} \frac{1}{2} & 0 & -\frac{1}{2} \\ 0 & 0 & 0 \\ -\frac{1}{2} & 0 & \frac{1}{2} \end{pmatrix} \tag{5}$$

The differential equation for $\rho(t)$ is given by

$$\frac{d\rho}{dt} = \frac{i}{\hbar}[\rho, \mathcal{H}] - \Gamma\rho + \dot{\rho}^o, \tag{6}$$

where $\mathcal{H} = -\vec{\mu}\cdot\vec{H}_o$, $\Gamma = 1/\tau$, the reciprocal lifetime of the excited state. The solution is

$$\rho_{mm'}(t) = \rho^o_{mm'} \exp[-i(m-m')\omega_o(t-t_o)] \exp[-\Gamma(t-t_o)], \tag{7}$$

where ω_o is the Larmor angular frequency, when the atoms are excited by sharply pulsed electron-impact excitation at $t = t_o$. Substitution of Eq. (7) into Eq. (3) yields

$$I(t) = I_o\Gamma \exp[-\Gamma(t-t_o)]\{A+B\cos[2\omega_o(t-t_o) + \phi]\}, \tag{8}$$

where A, B, and ϕ are time-independent constants showing the behavior of a damped harmonic oscillator -type function, and ω_o is the Larmor angular frequency.

Intensity Beat in Resonance Absorption

In a resonance absorption experiment, certain magnetic sublevels of the 7^3P_2 metastable state are coherently produced. The behavior of the metastable state was observed by monitoring the absorption of the linearly polarized resonance radiation. However, the electron beam was modulated as $(1+\cos\omega t)$. Therefore, the rate equation for $\rho_{mm'}(t)$ is given, from Eq. (6), by

$$\frac{d\rho_{mm'}}{dt} = \rho^o_{mm'}(1+\cos\omega t) - i(m-m')\omega_o\rho_{mm'} - \Gamma\rho_{mm'}. \tag{9}$$

The solution is given as

$$\rho_{mm'}(t) = \rho^o_{mm'}\left[\frac{1}{\Gamma+i(m-m')\omega_o} - \frac{\frac{1}{2}e^{i\omega t}}{\Gamma+i\omega+i(m-m')\omega_o} + \frac{\frac{1}{2}e^{-i\omega t}}{\Gamma+i\omega+i(m-m')\omega_o}\right], \tag{10}$$

where $\rho^o_{mm'}$ is given by Eq. (2). Substituting Eq. (10) into Eq. (4), when $|\mu\rangle = |^3P_2, \mu\rangle$ and $|m\rangle = |^3S_1, m\rangle$, in the case of the electric dipole transition from $7^3P_2 - 8^3S_1$ of Hg at $\lambda = 5461$ Å, we get

$$I(t) = A\left[\frac{1}{\Gamma} + \frac{2\cos(\omega t-\phi_o)}{\sqrt{\Gamma^2+\omega^2}}\right] + B\left[\frac{2\cos(\omega t-\phi_1)}{\sqrt{\Gamma^2+(\omega+2\omega_o)^2}} + \frac{2\cos(\omega t-\phi_2)}{\sqrt{\Gamma^2+(\omega-2\omega_o)^2}}\right] \tag{11}$$

for linearly polarized light. Here $\tan\phi_o = \frac{\omega}{\Gamma}$, $\tan\phi_1 = \frac{\omega+\omega_o}{\Gamma}$, and $\tan\phi_2 = \frac{\omega-2\omega_o}{\Gamma}$. Here A and B and x are time-independent constants that are dependent on the cross section of the excitation of the metastable state. The second term represents the intensity beat in the resonance absorption when the modulation frequency of the electron beam is equal to twice the Larmor frequency.

Experimental Results

Intensity Beats in Resonance Fluorescence

A fast-response Pierce-type electron gun with a Phillips cathode was constructed. With the energy near the threshold energy, the electron current was quite low, and an extremely low photon flux resulted. For this reason, a delayed coincidence counting technique was used. Well-distilled Cd metal was sealed into the envelope of the electron gun, which was enclosed in an oven with a window. A solution of nickel sulfate (14 g) and cobalt sulfate (10 g) in 100 cc of water was used to filter (Corning glass filter, type 7-54) the λ = 3261 Å resonance luminescence corresponding to the transition 7^3P_1 to 7^1S_0.

The λ = 3261 Å photons were detected through a filter and a Glan-Thompson quartz polarizer by a photomultiplier tube (RCA type 1P28). To observe the beat phenomena, we used a multichannel analyzer with a time-to-pulse-height converter. Figure 1 is the block diagram of the experimental apparatus. The arrival time of a photon with respect to the start of an electron beam pulse was measured. The measurements were repeated over and over at the repetition rate of 10 kHz until we obtained a high enough S/N ratio. Figure 2 shows the effect of beat at H_o = 0.88 G. The period of oscillation corresponds to 270 nsec, resulting in g_J = 1.5. The lifetime of $^3P_1$ Cd measured from the exponential decay is 2.2×10^{-6} sec. From Fig. 2, it is rather obvious that if the electron beam is pulsed at t = 0, $1/2\omega_o$, $1/\omega_o$, $2/2\omega_o$, $\cdots$, the damped oscillator-type function should become undamped. Such experiments were performed by Aleksandrov and Pebay-Peyroula.[6,7]

Intensity Beat in Resonance Absorption

The atom used to test our idea of the possibility of coherent excitation was the 6^3P_2 metastable state of Hg. Behavior of this state was observed by monitoring the absorption of linearly polarized λ = 5461 Å (6^3P_2-7^3S_1) resonance radiation propagating along the externally applied magnetic field. A high concentration of coherently excited metastable atoms was produced by means of a space-charge-neutralized high-flux low-energy electron-current flow. The experimental arrangement showing the methods of high-frequency modulation is shown in Fig. 3. We typically operated the electron tube at 3 MHz with 400 mA of peak-to-peak current at 100% modulation. Figure 4 shows the experimental result of the 2.89-MHz component of the optical absorption as a function of applied magnetic field. The peak absorption corresponds to a magnetic field of H = 688±10 mG at Larmor frequency ν_L = 1.445 MHz.

Conclusion

The advantage of these methods over the conventional double-resonance methods are

1) Any state can be excited by the electron impact, and
2) No rf magnetic field that perturbs the state is needed.

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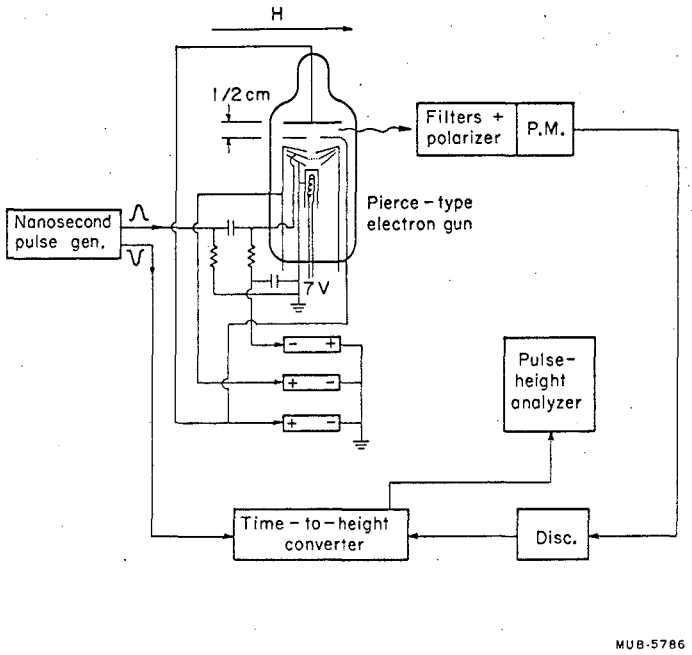

Fig. 1. Block diagram of light and beat experiment.

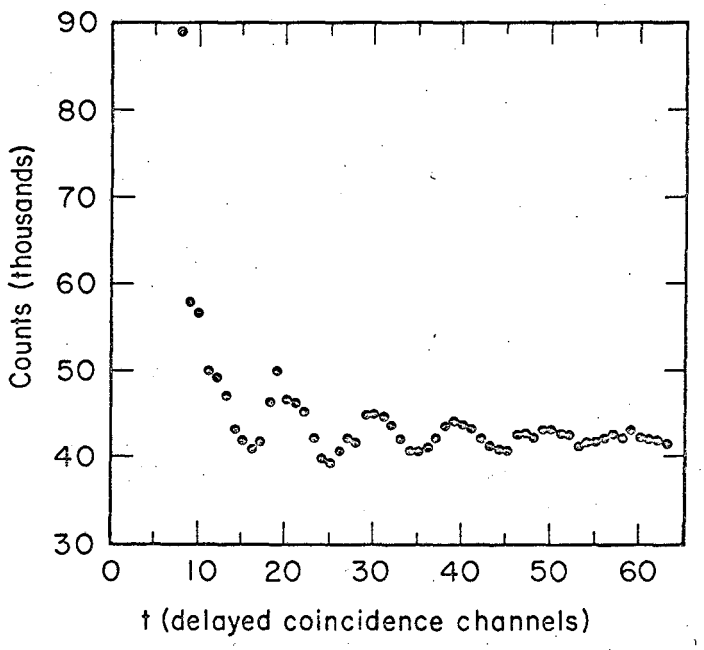

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Fig. 2. Quantum beats at 0.88 gauss.

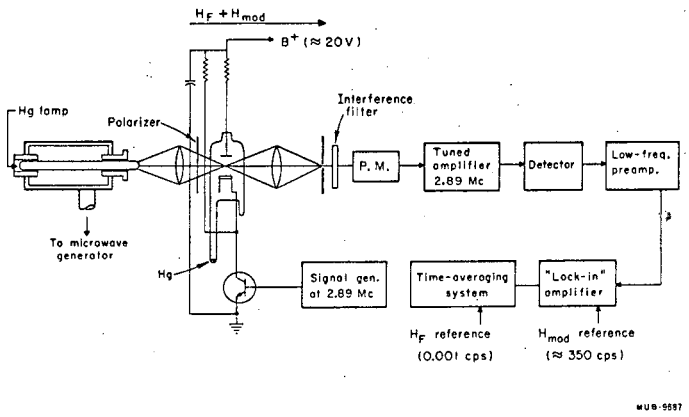

Fig. 3. Block diagram of the experimental apparatus of the resonance absorption beat experiment.

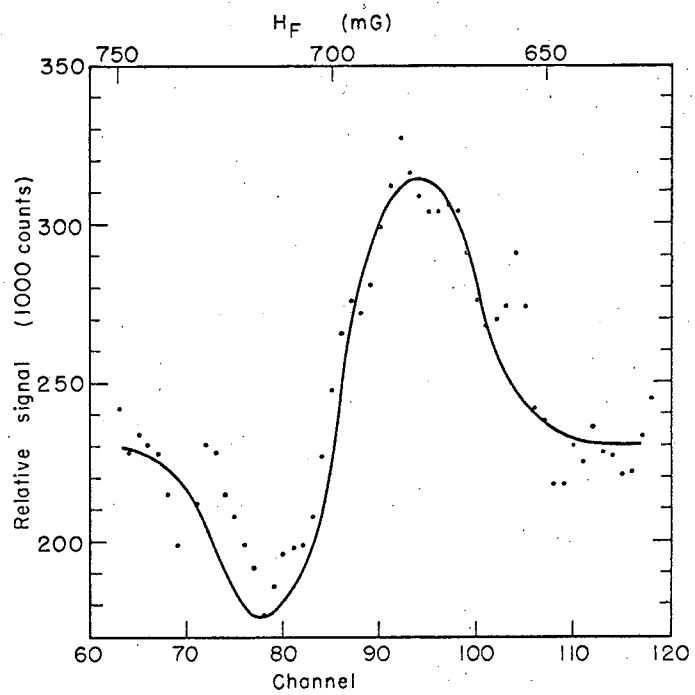

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Fig. 4. Resonance absorption beat signal with electron beam modulated at 2.89 MHz.

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