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## INDUCED ATOMIC CASCADE PROCESSES

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Polarization-direction correlation of incident and scattered photons in induced atomic absorption<sup>1</sup> which exhibit a second-order dependence on the inducing field are well known. Javan et al.<sup>2,3</sup> have recently reported similar correlations in induced cascade processes. In addition to these we have observed further correlations in induced atomic cascade processes which show a fourthorder dependence on the inducing field. An interesting feature of the fourth-order dependence is that amplitude modulation of the inducing field alters the time-averaged correlations.

The experiment to be described consisted of observing the dc magnetic field dependence of the spontaneous emission from the neon  $2p_4$  level under condition of multimode laser action between the  $3s_2$  and  $2p_4$  levels (0.633  $\mu$ ). A monochromator was used to select spontaneous emission from a given  $2p_4 - 1s$  transition emitted at right angles to the laser axis (see coordinate system in Fig. 1). The observed discharge region was placed in a weak variable magnetic field parallel to the laser axis, this region being kept small (~4 cm) to minimize magnetic field perturbation of the laser action. The earth's field was also cancelled out. An essential feature of the experiment was the insertion of two modulators in the cavity. One modulator, using acoustic standing wave diffraction,<sup>4</sup> was operated at the fundamental mode spacing frequency of the laser. Under this condition the laser spectrum consisted of approximately 26  $\text{TEM}_{00q}$  modes<sup>5</sup> with uniform spacings equal to (56.02±0.0001) Mc/sec, phase locked together to give an  $\approx 100\%$  amplitude-modulated field.<sup>6,7</sup> The other modulator, a mechanical chopper, modulated the laser action at audio frequencies to allow synchronous detection of the spontaneous emission dependent on induced emission. Oscillation at 3.39 microns was suppressed by introducing methane at one end of the cavity.

A calculation of the time-averaged secondorder scattering (induced emission  $3s_2 \rightarrow 2p_4$ , followed by spontaneous emission,  $2p_4 \rightarrow 1s_4$ ,  $1s_5$ ) was made by applying standard second-order theory<sup>1</sup> [Fig. 2(a)].<sup>8</sup> A caluclation was also made of the intensity of the 56-megacycle/second component of the spontaneous emission by applying the formalism for excitation by a modulated optical field<sup>9</sup> [Fig. 2(b)]. It should be noted that to second order the assumption of an amplitudemodulated exciting optical field does not affect the calculated time-averaged spontaneous emission [Fig. 2(a)].



FIG. 1. Coordinate system and neon level scheme.  $\vec{H}$  is the magnetic field,  $\vec{E}$  the linearly polarized laser field,  $\vec{k}_l$  the inducing field propagation vector, and  $\vec{k}_f$ the spontaneous emission propagation vector. Connecting lines show induced (straight line) and spontaneous (wavy line) transitions which contribute to the magnetic field-dependent part of the fourth-order process.

<sup>&</sup>lt;sup>3</sup>Anna Mani, Proc. Indian Acad. Sci. A20, 323 (1944).



FIG. 2. Theoretical and experimental plots of the spontaneous emission  $(2p_4 \rightarrow 1s_4)$  vs magnetic field. (a) Theoretical plot of the time-averaged spontaneous emission, including only second-order terms in E. (b) Theoretical plot of the 56-Mc/sec component of the spontaneous emission (assuming square law receiver response), including only second-order terms in E. (c) Experimental trace of the time-averaged spontaneous emission. (d) Experimental trace of the 56-Mc/sec component of the spontaneous emission. The letters x and z denote analyzer parallel to x or z, respectively. Thus the x traces correspond to  $\Delta m = \pm 1$ , the z traces to  $\Delta m = 0$  transitions. The theoretical signal amplitude has been rather arbitrarily normalized by setting calculated and experimental zero-field signal amplitudes equal.

Among the various differences in the experimental<sup>10</sup> [Figs. 2(c) and 2(d)] and theoretical curves [Figs. 2(a) and 2(b)], the existence of the dips in trace z of Fig. 2(c) is the most obvious. We suggest that these dips are the result of a fourth-order process involving the transitions shown in Fig. 1.<sup>11</sup> Three experimental observations can be cited in support of this view:

(1) As nearly as can be ascertained under the conditions of multimode laser operation, the dips show a fourth-power dependence on the amplitude of the maser field E.

(2) The dips were observed on the  $|2p_4m\rangle$  -  $|1s_4m\rangle$ , but not on the  $|2p_4m\rangle - |1s_5m\rangle$  transitions. This selective character of the effect is expected because of the forbidden  $|2p_40\rangle - |1s_50\rangle$  transition (Fig. 1).

(3) The dips at  $H \neq 0$  occur at magnetic field splittings of the  $3s_2$  state equal to integral multiples of the fundamental beat frequency f and depend strongly on the amplitude of the maser field modulation.

A calculation of the fourth-order contribution to the population density of the  $2p_4 m = 0$  state can be obtained by using density matrix formalism and perturbation theory after Lamb.<sup>12</sup> To include magnetic field dependence we extended Lamb's calculation to include two upper levels (magnetic sublevels of a single electronic state). We assume two-mode equal-amplitude (E) operation and stationary atoms. The fourth-order contribution to the population of the m = 0 state of the  $2p_4$  level has magnetic field-dependent terms (corresponding to the processes shown in Fig. 1) of the form

$$\rho_{bb}^{(4)} = -\frac{E^4}{16\hbar^4} \left( \frac{\Lambda_a}{\gamma_a} - \frac{\Lambda_b}{\gamma_b} \right) |\langle \alpha \mid \mu \rangle|^2 |\langle \alpha' \mid \mu \rangle|^2 \left\{ \frac{\beta_1}{\gamma_{ab}^{-2} (\gamma_a^{-2} + \Delta^2)} + \frac{\beta_2}{(\gamma_{ab}^{-2} + f^2/4) [\gamma_a^{-2} + (\Delta \pm f)^2]} + \cdots \right\}.$$

Here  $\langle \alpha | \mu \rangle$  is the matrix element for the electric dipole moment between upper state magnetic sublevel  $\alpha$  and lower state sublevel  $\mu$ ,  $\beta_1$  is a parameter of the order of 8 and depends on the relative positions of the laser modes and the atomic resonance,  $\gamma_a$  and  $\gamma_b$  are the decay constants for the  $3s_2$  and  $2p_4$  states respectively, and  $\gamma_{ab}$  $=\frac{1}{2}(\gamma_a + \gamma_b)$ . The rates of excitation to the  $3s_2$ and  $2p_4$  states per unit volume per unit time are denoted by  $\Lambda_a$  and  $\Lambda_b$  respectively. The symbol  $\Delta = 2g\mu_B H/h$ , where g,  $\mu_B$  and h are the g factor, Bohr magneton, and Planck's constant in that order. The symbol f represents the fundamental beat frequency of the laser. For many-mode operation, terms appear showing resonances at  $\Delta \pm 2f = 0$ ,  $\Delta \pm 3f = 0$ , etc. The correspondence of these calculated terms with the dips in trace z and even trace x of Fig. 2(c) is apparent. Oscillating terms (not shown) at f, 2f, etc., also occur. An incidental point of interest is that the width of the fourth-order Lorentzian resonance is given by  $\gamma_a$  permitting, for resolved fourthorder peaks, a measure of the upper state lifetime.<sup>13</sup>

We feel that one of the most striking features of this experiment is the contribution to the timeindependent scattering produced by amplitudemodulating the saturating field. Our two-mode calculation shows that the terms giving that contribution arise from the interaction of the same atom with both laser modes, the atom interacting with each mode twice. We suggest that an appropriate way of viewing such a process is to regard it as an interference of two time-dependent second-order processes which produces a timeindependent contribution to the saturated susceptibility.

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<sup>1</sup>See for example, P. A. Franken, Phys. Rev. 121.

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<sup>2</sup>A. Javan, Bull. Am. Phys. Soc. <u>9</u>, 489 (1964). <sup>3</sup>R. H. Cordover, A. Szöke, and A. Javan, Bull.

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<sup>6</sup>It should be noted that this condition is not typical of laser action, but is forced by the intracavity modula-tor.

<sup>7</sup>This estimate of the mode phase relations was made on the basis of the known optical spectrum and the output pulse shape (half-width  $\sim 2.5 \times 10^{-9}$  sec) as observed on a sampling oscilloscope.

<sup>8</sup>This calculation used a value for the  $2p_4$  state lifetime of  $0.67 \times 10^{-8}$  sec obtained by an independent measurement made at the same operating pressure (0.1 Torr <sup>20</sup>Ne and 0.7 Torr He) as the present experiment.

<sup>9</sup>A. Corney and G. W. Series, Proc. Phys. Soc. (London) <u>83</u>, 207 (1964).

<sup>10</sup>The second-order peak [Fig. 2(c)] provides a measure of the lower state lifetime; however, fourth-order processes also contribute to the peak half-width, the angular dependence of the ratio of second- and fourthorder contributions causing significant (e.g., 8%) variations in the apparent half-width as a function of observation angle. Valid lower state lifetime measurements thus require operation at low saturation levels or the addition of appropriate higher order corrections.

<sup>11</sup>A contribution to the magnetic field-dependent scattering from the process  $\langle m = \pm 1 | \rightarrow \langle m = 0 | \rightarrow \langle m = \pm 1 |$ is unique to a fourth-order induced event, and can be thought of as arising from the precession of the depletion-induced polarization of the upper (3s<sub>2</sub>) state. <sup>12</sup>W. Lamb, Phys. Rev. <u>134</u>, A1429 (1964). The authors are indebted to Professor Lamb for access to his manuscript prior to publication and for clarification of certain portions of that manuscript.

<sup>13</sup>We also note that deviations of theory and experiment remaining after accounting for higher order effects, particularly polarization discrepancies, should provide a measure of disorienting collision cross sections.

PHOTOPRODUCTION OF  $\pi$  MESONS BETWEEN 0.9 AND 4.0 BeV\*†

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We have measured protons and gamma rays in time coincidence using the apparatus shown schematically in Fig. 1. It consists of two rotating platforms copivoted about a liquid hydrogen tar-

get through which passes a photon bremstrahlung beam from an internal target in the Cambridge Electron Accelerator. The larger platform carries, in order, two quadrupoles (Q1 and Q2)