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[15]Our measurements show that the electron current was of the order of $10^{-3}$ of a 1-keV deuteron beam current which passed through the apparatus before cesium vapor or $H_2$ gas were admitted to the cells. Thus, if a $10^{-5}$-A deuteron current were available, we could expect a current of polarized electrons of about $10^{-8}$ A. Actually, a current of $10^{-5}$ A of deuterons is a conservative value for this estimate since J. L. McKibben and co-workers [Bull. Am. Phys. Soc. 12, 463 (1967)] measure beams of several hundred microamperes of 500-eV protons in a geometry similar to that needed for a source of polarized electrons.

# FREQUENCY SPECTRUM OF SPONTANEOUS AND STIMULATED LINE-NARROWING EFFECTS INDUCED BY LASER RADIATION*

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In an earlier Letter,[1] a laser-induced line-narrowing effect was utilized in a precise determination of isotope shifts in two Ne transitions. A recent paper[2] also reports observation of this effect and its application in studying some linewidth parameters in Ne. These experiments study fluorescence arising from the lower level of a Doppler-broadened laser transition to a third level. [See Fig. 1(a).] Viewed along the laser axis, the broad fluorescence line shape is dramatically influenced by the laser field. For a standing-wave field detuned from its atomic center frequency, the laser-induced change signals appear as resonant increases in intensity over two narrow intervals symmetrically located on opposite sides of the fluorescence center frequency. The fluorescence from the upper laser level [Fig. 1(c)], similarly viewed, would exhibit narrow resonant decreases in its over-all emission profile.

The over-all features of this effect may be described by noting that the standing-wave laser field selectively interacts with atoms whose velocities cause a Doppler shift of one of its traveling-wave components into resonance; this produces changes in the laser level populations—an increase in the lower level population and a decrease in the upper level population—over two narrow intervals symmetrically located about the center of the velocity distribution. A recent Letter[3] has analyzed the line-shape details for the cascade case [Fig. 1(a)] in terms of two-quantum transitions from level 2 to level 1, and predicts differing widths for the two laser-induced change signals. A similar line-shape asymmetry, described below, would appear in the change signals from the upper laser level [Fig. 1(c)]. Note that in the latter case, however, the $0 \rightarrow 1$ emission act is an inherently single-quantum event, re-

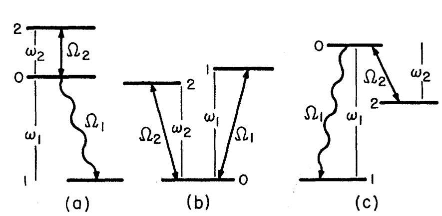


FIG. 1. Energy-level diagram.

quiring another description. The summary of a different treatment,[4] based on the density-matrix formalism, has described the spontaneous-emission profile in both cases. Line-shape behavior of similar origins has been encountered in the microwave region in experiments involving the interaction of two monochromatic, classical fields with a three-level system.[5,6] These considerations may be readily extended to the present case by including the Doppler effect, and noting that the spontaneous-emission spectrum from either laser level [level 0 of Figs. 1(a) and 1(c)] follows the emission line shape stimulated by a weak monochromatic field tuned through that resonance when the population of level 1 is ignored. In fact, recent experiments have studied the response of a Doppler-broadened three-level system coupled to two classical fields. The line-shape theory describing these experiments is directly applicable to important special cases of the effect observed in spontaneous emission. This Letter has three intimately related objectives: The first is to point out the direct relevance of the detailed analyses of Schlossberg and Javan[7] and Feld[8] to this problem; these also predict that one of the laser-induced resonances will be narrower than the other. Secondly, we emphasize that experiments based on stimulated versions of this effect demonstrate the different characteristics of the two laser-induced resonances. Thirdly, this Letter generalizes the theory in several ways, including important power-broadening effects.

The analysis of Ref. 7 describes the third-order interaction of two monochromatic fields with the folded Doppler-broadened system of Fig. 1(b) in which levels 1 and 2 are assumed to be closely spaced. Note that the analysis of a cascade system [Fig. 1(a)] in which the middle level lies about halfway between the other two levels follows identically. For purposes of illustration, let us assume that both transitions exhibit gain. Consider a strong standing-wave laser field $E_2(\Omega_2)$ detuned by an amount $|\Delta_2| > \gamma_{21}+\frac{1}{2}\gamma_0$ from $\omega_2$, the peak of the 2-0 Doppler response [Fig. 1(b)]; here $\gamma_{ij}=\frac{1}{2}(\gamma_i+\gamma_j)$, with $\gamma_j$ the natural decay rate of level $j$. As a weak monochromatic field $E_1(\Omega_1)$ is tuned through the 1-0 transition, it is shown that the broad Doppler-gain profile is considerably modified by the presence of $E_2$. The resulting line shape is obtained from Eqs. (33d) and (33e)[9] of Ref. 7:

$$P(\Omega_1)=G(\Omega_1)[1+\xi E_2^{0 2}\,\mathrm{Im}\{[(\Delta_2+\Delta_1)+i(\gamma_{21}+\gamma_0)]^{-1}+[(\Delta_2-\Delta_1)+i\gamma_{21}]\}],\quad \omega_1\approx\omega_2, \tag{1}$$

in which $\Delta_j=\Omega_j-\omega_j$, $\omega_j$ = atomic center frequency of $j$-0 transition, and $\Omega_j$ and $E_j^0$ are frequency and amplitude of $E_j(\Omega_j)$, respectively; $G(\Omega_1)$ is the linear Doppler response, a slowly varying function of $\Omega_1$, and $\xi$ is a constant factor. For $\Omega_2$ above $\omega_2$, (1) predicts narrow Lorentzian responses of widths $\Gamma_B=\gamma_1+\gamma_2+2\gamma_0$ below $\omega_1$ and $\Gamma_N=\gamma_1+\gamma_2$ above $\omega_1$, the latter being narrower by $2\gamma_0$ and independent of $\gamma_0$. In subsequent discussion we shall refer to the "broad" ($\Gamma_B$) and "narrow" ($\Gamma_N$) resonances, respectively.

The narrow resonance, $\Gamma_N$, predicted by (1) has been observed and fully verified[10] for $\Gamma_N \ll \Gamma_B$. In these experiments levels 1 and 2 are tunable Zeeman components of an upper laser level connected to a common lower level, 0. The monochromatic fields are two oscillating laser modes of fixed frequencies determined by the cavity length. The effect is observed as sharp decreases in the laser output as the 2, 1 level splitting is magnetically tuned. These methods, however, have not been applied to the observation of $\Gamma_B$, which requires absolute frequency control as in Lamb-dip experiments (see Ref. 7).

We report here an extraordinary manifestation of the broad resonance, observed in high-resolution studies of the $3p\,^3P_{0,2,1}$-$3s\,^3S_1$ atomic-oxygen fine-structure laser oscillations at 8446 Å. The relevant fine-structure components of this system consist of two resolved and closely spaced Doppler-broadened transitions forming folded three-level systems of the type shown in Fig. 1(b). For reasons unrelated to the present discussion,[11] (i) the central portion of each of the fine-structure gain profiles is entirely depleted and appreciable gain exists only on the wings, and (ii) laser action most readily occurs on the high-frequency side of the strongest (2-0) fine-structure component. For each laser mode oscillating on the 2-0 transition, two Lorentzian holes of enormously different widths selectively deplete the 1-0 Doppler gain profile; the low-frequency hole, of width $2\gamma_0$

$+\gamma_1+\gamma_2=130$ MHz, is about 15 times broader than the high-frequency hole, of width $\gamma_1+\gamma_2=9$ MHz.[12] Because of multimoding on the 2-0 transition, multiple hole pairs are burnt into the 1-0 transition. The broad low-frequency holes overlap, completely suppressing laser action below the 1-0 line center, and oscillation can occur between the narrow, nonoverlapping holes above the center frequency. The 8446-Å spontaneous emission and laser output have been studied photographically with a high-resolution Fabry-Perot interferometer with free spectral range of 0.8 $\mathrm{cm}^{-1}$ under a wide range of conditions. The laser oscillations were obtained in argon-oxygen and neon-oxygen mixtures,[13] using a 3-m cavity with 50-MHz mode spacing. The operating pressure was kept below 1 Torr to minimize pressure-broadening effects. To establish the frequency shifts, Fabry-Perot images of the laser oscillations and the spontaneous emission were superimposed upon the same glass plate emulsion. In absence of laser oscillation, the spontaneous-emission analysis shows the following in order of increasing frequency: a well-resolved fine-structure component (1-0) with a completely symmetrical profile, and the strongest fine-structure component (2-0), overlapped on the high-frequency wing by the third (weak) fine-structure component. Close to threshold, laser oscillation first occurs on the high-frequency wing of the 2-0 component, where maximum gain occurs, as explained in Ref. 11 (ii). Further above threshold the 1-0 transition also breaks into oscillation. Despite the observed symmetry of the 1-0 profile, this concurrent oscillation occurs above the 1-0 center frequency, an effect which must follow from the depletion of gain over the entire low-frequency wing, as described above. (It must also be pointed out that in the observations reported here the 1-0 oscillation was at all times close to threshold and considerably weaker than the 2-0 oscillation.[14])

Reference 7 treats the field interactions up to third order. We now outline a different approach,[8] formulated in terms of single- and double-quantum transitions, which includes power-broadening effects due to a strong laser field. The latter are of considerable interest: On the theoretical side, it is important to inspect whether or not the strikingly simple line-shape behavior is merely characteristic of a third-order calculation; on the experimental side, it is important to know the influence of power broadening on the observed line shape. Consider the cascade system of Fig. 1(a). [The final result will be written in a form also valid for emission from the upper laser level, Fig. 1(c).] It is desired to calculate the 0-1 emission spectrum stimulated by the weak traveling-wave field $E_1(\Omega_1)$ in the presence of the strong standing-wave field $E_2(\Omega_2)$ coupled to the 2-0 transition. Specifically, standing-wave effects are avoided by taking $\Omega_2$ detuned ($|\Delta_2|>\gamma_{20}$); then oppositely propagating traveling-wave components of $E_2$ do not couple, and the interaction consists of these components independently coupled with $E_1$.

Consider an ensemble of atoms moving with given axial velocity $v$, the $+z$ axis being defined by the propagation direction of the traveling-wave field $E_1$. In the atoms' rest frame the incident fields appear Doppler-shifted to frequencies $\Omega_1'=\Omega_1(1-v/c)$ and $\Omega_2'=\Omega_2(1-\epsilon v/c)$ in which $\epsilon=\pm1$ indicates the traveling-wave component of $E_2$ propagating along the $\pm z$ direction, respectively. We now require a solution in the atoms' rest frame in which the three-level system is coupled to one of the traveling-wave components of the strong field $E_2$ at frequency $\Omega_2'\sim\omega_2$ and to the weak traveling-wave field $E_1$ at frequency $\Omega_1'\sim\omega_1$. The resonant interaction of two monochromatic fields with a three-level system was treated in Ref. 5 for the case of $\gamma_j$ all equal. The perturbation method consisted of first obtaining a closed-form solution to the Schrödinger equation for $E_1^0=0$ and $E_2^0$ arbitrary, and then using this result to generate a solution valid to first order in $E_1^0$. When the method is extended to the case of arbitrary $\gamma_j$,[15] the emitted power induced by $E_1$ at $\Omega_1'$ is

$$8\hbar\Omega_1'|\beta_1|^2\,\mathrm{Im}\left[n_2|\beta_2|^2\frac{[L_2-2(\gamma_{20}/\gamma_0)R]}{AB}+n_1\frac{R}{B}-n_0\left\{\frac{R}{B}+|\beta_2|^2\frac{[L_2-2(\gamma_{20}/\gamma_0)R]}{AB}\right\}\right]. \tag{2}$$

Here, $A=|L_2|^2+(4\gamma_{20}^2/\gamma_0\gamma_2)|\beta_2|^2$ and $B=-RL_1^*+|\beta_2|^2$, with $L_j=(\Omega_j'-\omega_j)+i\gamma_{j0}$, $R=[(\Omega_1'+\Omega_2')-(\omega_1+\omega_2)]-i\gamma_{21}$, $|\beta_j|=|\mu_{j0}E_j^0/4\hbar|$, and $\mu_{j0}$ is the electric-dipole matrix element connecting levels $j$ and 0; $n_j$ is the number of background atoms with velocity $v$ in level $j$, i.e., the population in absence of the strong laser field. In Eq. (2) the $n_2$ coefficient is obtained from the $2 \to 1$ transition rate due to double-quantum emission at $\Omega_1'$ and $\Omega_2'$; the $n_0$ coefficient, in contrast, is obtained from the single-quantum emission rate arising from $0 \to 1$ transitions as modified by the presence of $E_2$; the $n_1$ coefficient results from the reverse processes, namely, double-quantum $1 \to 2$ transitions and single-quantum $1 \to 0$ transitions[16] (see Ref. 5). As a check of the detailed algebra, Eq. (2) has also been obtained in an independent calculation using the ensemble-averaged density matrix to estimate the induced polarization at $\Omega_1'$, an approach equivalent to the one presented here.[8]

It is important to note that (2) is entirely valid for the case in which the $\gamma_j$'s are interpreted as decay rates arising from hard collisions. The detailed features of the line shape predicted[5] by (2) for equal $\gamma_j$ have been fully verified in the microwave region where Doppler effect is negligible and the linewidths are entirely due to collision effects.[6]

The complete emission spectrum is obtained by averaging (2) over the atomic velocity distribution for $\epsilon = +1$ and for $\epsilon = -1$. In the fully Doppler-broadened limit $\gamma/D \ll 1$ ($\gamma \sim \gamma_{ij}$ and $D$ = Doppler width), and for $\omega_1 \geq \omega_2$,

$$P(\Omega_1) = G(\Omega_1)\left[1 + \frac{N_0 - N_2}{N_0 - N_1}\xi' E_2^{02}\,\mathrm{Im}\left\{\left[\left(\Delta_1 + \sigma\frac{\omega_1}{\omega_2}\Delta_2\right) + i\frac{\Gamma_B}{2}\right]^{-1} + \left[\left(\Delta_1 - \sigma\frac{\omega_1}{\omega_2}\Delta_2\right) + i\frac{\Gamma_N}{2}\right]^{-1}\right\}\right], \tag{3}$$

in which $\frac{1}{2}\Gamma_B = \gamma_{10} + (\omega_1/\omega_2)\gamma_{20}Q + \frac{1}{2}\gamma_0(Q-1)$ and $\frac{1}{2}\Gamma_N = \gamma_{10} + (\omega_1/\omega_2)\gamma_{20}Q - \frac{1}{2}\gamma_0(Q+1)$; $N_j$ is the total background population of level $j$, $\xi'$ is a proportionality factor $>0$, $Q^2 = 1 + 4|\beta_2|^2/\gamma_0\gamma_2$, and $G(\Omega_1) \propto (N_0 - N_1)(E_1^0)^2$ is the power emitted at $\Omega_1$ by the Doppler-broadened 0-1 transition induced by $E_1$ in the absence of the laser field. Equation (3) shows the power broadening influence of the laser field, which enters in a remarkably simple way. Equation (3) has been written in a form valid for both cascade ($\sigma = -1$) and folded ($\sigma = +1$) cases, Figs. 1(a) and 1(c).[17]

The discussions of Ref. 3 are consistent with the weak-field limit of our treatment for the case of $N_0 = 0$. Note, for instance, that for $|\beta_2|^2 \ll 1$ and in the limit of $\gamma_{21} \to 0$, the frequency dependence of the $2 \to 1$ transition rate, obtained from the $n_2$ coefficient of our Eq. (2), would involve a $\delta$ function, becoming $\delta(\Omega_1' + \Omega_2' - \omega_1 - \omega_2)|L_2|^{-2}$; the $\gamma_{21} = 0$ discussion of Ref. 3 is equivalent to averaging this distribution over velocities.

To emphasize the significance of the role played by $N_0$, the background atoms in level 0, consider a cascade system in which only level 0 is populated (i.e., $N_1 = N_2 = 0$). Then in the rest frame of an atom an applied laser field at $\Omega_2'$ will diminish the transition rate at $\Omega_1'$, leading to two holes of width $\Gamma_B$ and $\Gamma_N$ superimposed upon the emission profile [see Eq. (3)]. As discussed earlier, a $0 \to 1$ transition is an inherently single-quantum event[5] and may not be described in terms of a double-quantum process as in a $2 \to 1$ transition.

As pointed out above, in the oxygen[10,18] and xenon[10] experiments, $\Gamma_N$ and $\Gamma_B$ differ enormously. In the Ne spontaneous-emission experiments reported in Refs. 1 and 2, however, they are expected to differ by only about 30%. The observation of this difference would require high-finesse Fabry-Perot analysis and good laser stability, and has not yet been achieved.

In averaging Eq. (2) for the case of finite $\gamma_{21}$, a number of cancellations occur in the fully Doppler-broadened limit ($\gamma/D \ll 1$), leading to a particularly simple expression. It is important to point out that such cancellations do not occur in higher orders of $\gamma/D$. For instance, the complete calcellation of $\gamma_0$ in $\Gamma_N$, which occurs in the case of $\omega_1/\omega_2 \approx 1$, will not occur in the next order of $\gamma/D$.

A paper including complete algebraic details and additional discussions will be published elsewhere (Feld and Javan[8]).

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[9]There are several relevant misprints in Eq. (33): In line (33e), $E_1E_2$ should read $E_1E_2^2$; line (33d) should read $+(|\mu_{12}\mu_{31}|^2/\gamma_1)(N_3-N_1)\frac{1}{2}E_1E_2^2[\gamma-i(\omega_B-\nu_B)]^{-1}$.

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[11]For explanation of (i), see Ref. 7 and M. S. Feld, B. J. Feldman, and A. Javan, Bull. Am. Phys. Soc. 12, 669 (1967), and "Frequency Shifts of the Fine Structure Oscillations of the 8446-Å Atomic Oxygen Laser," to be published; (ii) is merely due to the presence of the weak fine-structure component with gain, which overlaps the high-frequency wing of the 2-0 transition ($^3P_2$-$^3S_1$). The 1-0 transition ($^3P_1$-$^3S_1$), however, is completely symmetrical and free of overlap.

[12]For oxygen linewidths, see W. L. Wiese, M. W. Smith, and B. M. Glennon, Atomic Transition Probabilities, U. S. National Bureau of Standards National Standard Reference Data Series–4 (U. S. Government Printing Office, Washington, D. C., 1966), Vol. 1.

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[14]A study of the intensity of the 1-0 laser oscillations as a function of cavity length would be of interest.

[15]In this case the time-dependent wave function $\Psi$ is obtained from a three-level Schrödinger equation to which radiative decay terms have been added. For

$$\Psi = \sum_{j=0}^{4} c_j \exp(-iW_j t) u_j,$$

with $u_j$ the eigenfunction of level $j$ of energy $\hbar W_j$, the coupled equations are $\dot{c}_i + \sum_j (a_{ij} - \frac{1}{2}\gamma_j \delta_{ij}) c_j$, in which $a_{ij} = -[\mu_{ij} E(t)/i\hbar] \exp[i(W_i - W_j)t]$, and $E(t)$ is the sum of the two traveling-wave fields as seen in the atoms' rest frame.

[16]As an example, for an atom in level 0 at initial time $t_0$, $|c_j(t=t_0, t_0)| = \delta_{j0}$ and the $0 \to 1$ transition rate at a later time $t$ is $\gamma_1 |c_1(t,t_0)|^2$ (see preceding footnote). Thus, the total stimulated power emitted by background atoms in level 0 is $\hbar\Omega_1' n_0 \gamma_0 \gamma_1 \int_{-\infty}^{t} |c_1(t,t_0)|^2 dt_0$.

[17]In extending Eq. (2) to the spontaneous-emission case, the population of level 1, $n_1$, should be set equal to 0 and the energy density of the weak probe field, $(E_1^0)^2/8\pi$, should be replaced by $(\hbar\Omega_1'^3/8\pi^3c^3)d\Omega_1' dS$, where frequency interval $d\Omega_1' \ll \gamma$ and $dS$ is a small solid angle in the forward direction ($+z$ axis); $E_2$, the laser field, remains in its classical monochromatic form. Similar remarks apply to Eq. (3); note, in particular, that $G(\Omega_1)$ becomes the usual Doppler-broadened spectrum of the power emitted spontaneously into $d\Omega_1 dS$ with given polarization.

[18]See Feld, Feldman, and Javan, Ref. 11.

# EVIDENCE FOR A NEW KIND OF ENERGETIC NEUTRAL EXCITATION IN SUPERFLUID HELIUM*

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We present evidence showing that neutral excitations which are not photons are produced in superfluid liquid helium in the presence of a $Po^{210}$ alpha-particle source. At low temperatures these excitations travel through the liquid with negligible scattering. They have sufficient energy to generate positive ions and electrons at the surface of the liquid or at a suitable metal plate immersed in the liquid.

Charged particles introduced into superfluid liquid helium form ion complexes which have been used to study scattering effects due to rotons, phonons, and $He^3$ atoms.[1,2] Such charged particles have also been used to produce quantized vortex rings[3] and to investigate the properties of such rings as well as of vortex lines.[4] In this Letter we report evidence for the existence of energetic neutral excitations in He II and a study of the emergence of charged particles through the surface of the liquid at temperatures below 1°K.

Our experimental arrangement, illustrated in Fig. 1, is similar to that of Rayfield and Reif.[3] The ion source $S$, plated with $Po^{210}$, is immersed in liquid helium; it emits alpha particles which generate ions within a 0.2-mm thick layer of liquid adjacent to $S$. Electric potentials in the apparatus are adjusted by means of metal grids. The current arriving at the collector $C$ is measured with a vibrating-reed electrometer. All the electrodes are gold plated to avoid formation of insulating oxide layers. At the temperatures below 0.6°K used in most of our experiments, the helium vapor pressure is so low that atomic mean free paths in the vapor above the surface of the liquid are long compared to the dimensions of the apparatus; hence the vapor region is effectively a vacuum.

Consider the experimental situation of Fig.