PHYSICAL REVIEW VOLUME 140, NUMBER 2A 18 OCTOBER 1965

Theory of Laser Cascades

H. HAKEN
Institut für Theoretische und Angewandte Physik, Technische Hochschule, Stuttgart, Germany

AND

R. DER AGOBIAN AND M. PAUTHIER
Laboratoire Central de Télécommunications, Paris, France

(Received 5 April 1965)

Two- and three-step laser cascades have been recently detected experimentally in He-Ne mixtures and in pure neon. In the present paper a two-step cascade is treated in detail theoretically, starting with the density-matrix formulation. The corresponding equations can be simplified to rate equations under an assumption about the relative size of certain phase-memory times, which seems well justified in the present case. Because there are usually only minor differences in the results between a gaseous laser and a solid-state laser with an inhomogeneously broadened line, and because the mathematical treatment of the gas laser is more involved, the solid-state case will be treated here. The laser condition for both the lower and upper transitions is derived for the most general case, i.e., that all three levels are pumped. Finally, a special two-step cascade is treated in detail. The results are in qualitative agreement with the experimental findings, although a quantitative comparison is still impeded by the lack of an exact knowledge of some of the parameters, especially the radiative linewidth.

1. INTRODUCTION

RECENTLY, the detection of laser cascades has been announced in a series of papers.[1–11] Although these cascades were first observed in a mixture of He and Ne, similar phenomena have also been detected in a pure gas system.[12] The experimental facts can be summarized in the following way: Let us consider a gas laser with a wide-band cavity, the active medium consisting of a discharge in a mixture of He and Ne. Under standard conditions, for example $p_{\text{tot}}=0.6$ Torr with a ratio He/Ne = 4, several lines are simultaneously present in the spectrum of this source. The accurate determination of the wavelengths of these lines and the subsequent identification of the transitions responsible for the light emitted shows that besides $2p$-$2s$ and $3p$-$3s$ transitions we have some $2s$-$3p$ transitions simultaneously present as, for example, those which can be seen on the energy-level diagram of the Fig. 1. It is clearly shown in this figure that the transitions are grouped; each element of one group is related to the others by optical connection. For example, in the group consisting of the $3p_4$-$3s_2$, $2s_2$-$3p_4$, and $2p_4$-$2s_2$ transitions, the suppression of the upper transition ($\lambda=3.39\ \mu$) by insertion of an absorptive medium (e.g., methane) in the cavity simultaneously eliminates the 2.395-μ line and reduces noticeably the intensity of the 1.15-μ line. The study of the variations of intensity of these lines with pressure shows that the 1.15-μ line has 2 maxima, the first being in close coincidence with the maximum of the 2.395-μ line and the other in the near vicinity of the maximum of the 3.39-μ line. The suppression of the 3.39-μ line eliminates, besides the 2.395-μ line, the first maximum of the 1.15-μ line but does not alter noticeably the other (Fig. 2). Moreover the 2.395-μ line shows absorption in the same medium ($\beta\simeq 7$ dB/m) when the cavity is removed. From these facts, it results that this last line is optically pumped by the 3.39-μ line and besides that we have a transmission of the pumping process to the 1.15-μ line (suppression of one maximum with methane).

In a He-Ne mixture the main population mechanism of the $3s_2$ level can be attributed to the reaction

$$\text{He}(2^1S)+\text{Ne}\rightarrow\text{He}+\text{Ne}(3s_2)+\Delta E_1$$

after Benton *et al.*[13] For the $2s_2$ level we have

$$\text{He}(2^3S)+\text{Ne}\rightarrow\text{He}+\text{Ne}(2s_2)+\Delta E_2$$

as pointed out by Javan *et al.*[14] In a wide-band optical cavity the large cross section of the first reaction ($\sigma\simeq 4\times10^{-16}$ cm^2) leads to lower-level pumping through a cascading-stimulated emission following the scheme:

$$3s_2\rightarrow 3p_4\rightarrow 2s_2\rightarrow 2p_4\,.$$

Although this cascade is not always the main populating

---

[1] R. A. McFarlane, W. L. Lamb, C. K. N. Patel, and C. G. B. Garrett, *Third International Conference on Quantum Electronics*, edited by N. Bloembergen and P. Grivet (Dunod Cie., Paris, 1963).
[2] R. Cagnard, R. Der Agobian, R. Echard, and J. L. Otto, Compt. Rend. **257**, 1044 (1963).
[3] R. Der Agobian, J. L. Otto, R. Echard, and R. Cagnard, Compt. Rend. **257**, 3844 (1963).
[4] H. J. Gerritsen and P. V. Goedertier, Appl. Phys. Letters **4**, 20 (1964).
[5] R. Grudzinski, M. Pailette, and J. Becrelle, Compt. Rend. **258**, 1452 (1964).
[6] J. L. Otto, R. Cagnard, R. Echard, and R. Der Agobian, Compt. Rend. **258**, 2779 (1964).
[7] W. L. Faust, R. A. McFarlane, C. K. N. Patel, and C. G. B. Garrett, Phys. Rev. **133**, A1476 (1964).
[8] R. Der Agobian, R. Cagnard, R. Echard, and J. L. Otto, Compt. Rend. **258**, 3661 (1964).
[9] W. R. Bennett, Jr., Bull. Am. Phys. Soc. **9**, 500 (1964).
[10] V. N. Smiley, Appl. Phys. Letters **4**, 123 (1964).
[11] R. Der Agobian, J. L. Otto, R. Cagnard, and R. Echard, Compt. Rend. **259**, 858 (1964).
[12] R. Der Agobian, J. L. Otto, R. Cagnard, and R. Echard, Compt. Rend. **259**, 323 (1964).
[13] E. E. Benton, E. E. Ferguson, F. A. Matson, and W. W. Robertson, Phys. Rev. **126**, 206 (1962).
[14] A. Javan, W. R. Bennett, Jr., and D. R. Herriott, Phys. Rev. Letters **6**, 106 (1961).

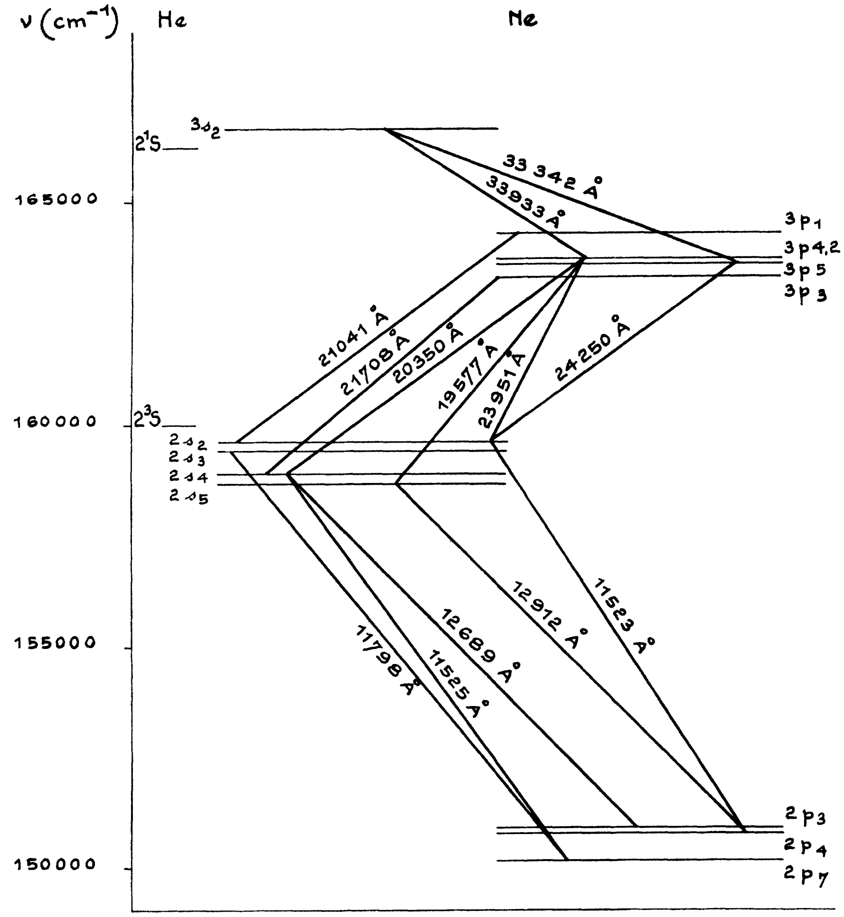

FIG. 1. Energy-level diagram showing the different cascades of neon generated by second-kind collisions of neon atoms with $2\,^1S$ He atoms.

process for these lower levels, it cannot be neglected. Let us mention also the case of another cascade observed in the same mixture for which the intermediate transition is not usually suppressed by eliminating the upper transition but shows only an important decrease in its intensity.

It is

$$3s_2 \rightarrow 3p_4 \rightarrow 2s_4 \rightarrow 2p_3\,.$$

In this case the lower transition is probably not very efficiently pumped by the action of the He metastable because the $2s_4$ level is energetically far ($\simeq$0.15 eV) away from the 2^3S level; the action of the He in the absence of the upper transition is probably to increase the temperature of the electron gas. For this cascade the cascade pumping process is much more important for the lower transition than in the first case since it is possible to find conditions (for example by shortening the discharge length) in which we suppress it by blocking the 3.39-μ line.

Similar effects have also been observed in pure neon. For example, the simultaneous observation in Ne of two lines in stimulated emission, namely the 2.10-μ line (transition $2s_2$-$3p_1$) and the 1.15-μ line (transition $2p_4$-$2s_2$), with a higher gain for the upper transition and an appreciable reduction in the gain of the lower transition by use of dielectric mirrors centered at 1.15 μ demonstrates experimentally the efficiency of a cascade pumping process.

In the present paper we intend to develop a theory of the laser cascade in order to elucidate among others the following questions:

(1) What requirements are to be fulfilled in general for a laser cascade to occur and what are the critical parameters?

(2) In what way does the lower transition depend on the upper one? In particular, is there a conservation of phase?

In order to treat these and related questions we use the density-matrix formalism. We confine ourselves to a two-step cascade between three optical levels, which can be pumped from a ground level or a set of levels independent of the levels participating in the laser process. The generalization to more complicated cases does not introduce supplementary theoretical difficulties, but leads to very lengthy formulas. Although all experiments showing laser cascades have been performed with gas lasers, it is not unreasonable to believe that similar results can be obtained with solid-state lasers. In an earlier paper[15] the close analogy between the theoretical

[15] H. Haken and H. Sauermann, Z. Physik **176**, 47 (1963).

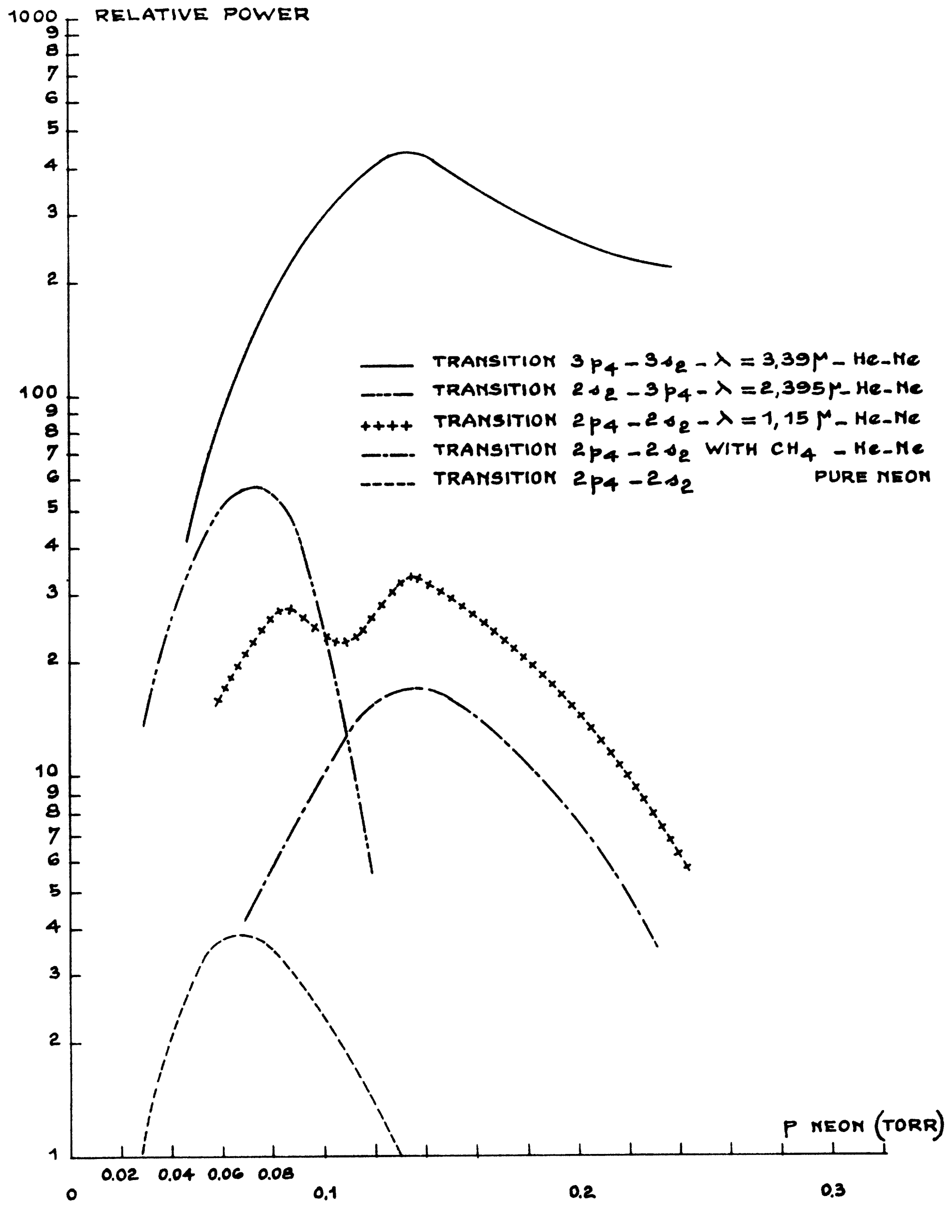

FIG. 2. Compared intensities of the different transitions of a neon cascade. Quenching the first maximum of the 1.15-μ line with CH_4 clearly shows the contribution of the cascade to the populating of the $2s_2$ level.

treatment of a solid-state laser and that of a gaseous laser has been demonstrated. Since the case of fixed atoms is mathematically somewhat simpler, this case is treated in the present paper. For the gas laser, the results are not fundamentally different, the corresponding calculations have been performed and will be published later.

The cooperation of two fields has been extensively investigated in the case of a three-level maser by Javan, Lamb, and Wilcox and others.[16–18] Our present investigation requires extension of these maser results in several directions: as mentioned above, our system should consist of at least four levels; the wavelength should be small compared to the dimension of the cavity; and the theory should allow for both homogeneously and inhomogeneously broadened lines.

2. HAMILTONIAN AND EQUATIONS OF MOTION

In this section we derive a Hamiltonian for the general case of a solid-state laser. We assume that the laser consists of a set of impurity atoms embedded in a solid matrix. It will be assumed that these impurity atoms are so far apart that there is no direct interaction between

[16] A. Javan, Phys. Rev. **107**, 1579 (1957).

[17] L. R. Wilcox and W. E. Lamb, Jr., Phys. Rev. **119**, 1915 (1960).

[18] N. Bloembergen and Y. R. Shen, Phys. Rev. **133**, A37 (1964). Further references are given here.

their electrons, for instance by Coulomb interaction or dipole-dipole coupling. Each of these atoms is supposed to possess three optically active levels and a fourth one which acts as a reservoir. The pumping process is supposed to be incoherent, for instance, provided by an incoherent light source. For gas atoms one can make similar assumptions in a series of cases (low pressure, etc.): the atoms participating in the lower process do not interact (collide) during their single light-emitting phase; the only important perturbation comes from "pumping" collisions.

As usual, we assume that there exists a set of decaying light modes, which are distinguished by their spatial behavior, their frequencies and their lifetimes. Since, for moderate pumping, only modes with the longest lifetime can participate in the laser process, we may confine ourselves to those. Furthermore, only those modes which are close to the resonance frequency of optical transition have sufficient gain. Therefore we may consider explicitly only three modes, or, if the 1-3 transition is forbidden, only the two modes corresponding to the transition $1 \to 2$ and $2 \to 3$. Whereas for a homogeneously broadened line (and running waves) this procedure is perfectly all right, an inhomogeneous transition might support several modes, their frequencies being sufficiently close to the center of the atomic line. In Sec. 4 we will briefly indicate how this effect can be taken into account.

We start with the formalism of second quantization which describes the annihilation and creation of electrons and light quanta. We denote the creation operator for an electron at atom μ in the state i by $a_{i,\mu}^\dagger$ and the corresponding annihilation operator by $a_{i,\mu}$. The energy of the state i is given by $\hbar\epsilon_i$. The creation operator for a light quantum of mode λ is denoted by $b_\lambda^\dagger$ and the corresponding annihilation operator by b_λ. Making use of the fact that the occupation number is given by

$$N_{i,\mu}=a_{i,\mu}^\dagger a_{i,\mu}\,,$$

we can write the energy of the electronic levels in the form

$$H_{\mathrm{EL}}=\sum_\mu \sum_{i=1}^{3} \hbar\epsilon_i a_{i,\mu}^\dagger a_{i,\mu}\,. \tag{2.1}$$

The energy expression of the light field takes the well-known form

$$H_L=\hbar\omega_1 b_1^\dagger b_1+\hbar\omega_2 b_2^\dagger b_2\,. \tag{2.2}$$

The interaction between light field and electrons is described by terms in which one photon is either annihilated or created while one electron is thrown out of one state i into another state j. This process can be described by

$$a_{j,\mu}^\dagger a_{i,\mu} b_\lambda$$

for light absorption, and by

$$a_{j,\mu}^\dagger a_{i,\mu} b_\lambda^\dagger$$

for light emission.

The total interaction Hamiltonian then reads for the laser cascade

$$H_I=\hbar\sum_\mu\{a_{1,\mu}^\dagger a_{2,\mu}h_{12,\mu}(b_1^\dagger+b_1)+a_{2,\mu}^\dagger a_{1,\mu}h_{12,\mu}^*(b_1+b_1^\dagger)$$
$$+a_{2,\mu}^\dagger a_{3,\mu}h_{23,\mu}(b_2^\dagger+b_2)+a_{3,\mu}^\dagger a_{2,\mu}h_{23,\mu}^*(b_2+b_2^\dagger)\}\,. \tag{2.3}$$

The coefficients $h_{ij,\mu}$ are given (in the dipole approximation) by

$$h_{ij,\mu}=G_{ij}(8/V)^{1/2}\sin(k_xX_\mu)\sin(k_yY_\mu)\sin(k_zZ_\mu)\,, \tag{2.4}$$

where

$$G_{ij}=-(e/m)[2\pi/\hbar\omega_\lambda]^{1/2}\langle\varphi_i\mathbf{p}\varphi_j\rangle\cdot\mathbf{e}_\lambda\,. \tag{2.5}$$

$\langle\varphi_i\mathbf{p}\varphi_j\rangle$ is the optical matrix element of a single atom for the corresponding transition. $\mathbf{k}$ is the wave vector of the light field and V is the volume of the cavity. $\mathbf{e}_\lambda$ denotes the polarization of the light wave λ. The total Hamiltonian reads

$$H=H_{\mathrm{EL}}+H_L+H_I\,, \tag{2.6}$$

where H_{EL}, H_L and H_I are given by (2.1), (2.2), and (2.3), respectively. In the following we neglect, as usual, terms which describe photon creation together with an upward electronic transition or photon annihilation together with a downward electronic transition, because these terms are antiresonant.

The most elegant way to deal with our problem is to derive the equations of motion in the Heisenberg picture, which means that we assume the operators depend explicitly on time. For this end we proceed in two steps. First we derive the equations of motion for operators of the form $a_i^\dagger a_j$ using as usual

$$(a_i^\dagger a_j)^{\cdot}=(i/\hbar)[H,a_i^\dagger a_j]\equiv(i/\hbar)(Ha_i^\dagger a_j-a_i^\dagger a_jH)\,, \tag{2.7}$$

where H is the total Hamiltonian (2.6). However, this Hamiltonian contains neither the atomic decay into the nonlasing modes, nor the atomic decay by radiationless processes, nor the pumping process. Therefore, in a second step we take these effects into account by applying appropriate heat baths to the system (2.6). For the treatment of this problem we adopt the method of Wangsness and Bloch[19] which was developed for the problem of spin resonance, similar in form, and apply the following procedure[20]: We take expectation values of both sides of (2.7) with respect to a superposition of atomic states and take the trace over the heat baths:

$$\rho_{ij,\mu}=\langle a_{i,\mu}^\dagger a_{j,\mu}\rangle\,. \tag{2.8}$$

Simultaneously, we introduce loss terms as caused by the heat baths. Because they enter into the equations of

[19] R. K. Wangsness and F. Bloch, Phys. Rev. **89**, 728 (1953).

[20] A most explicit derivation using heat baths with different temperatures has recently been given by W. Weidlich and F. Haake, Z. Physik **185**, 30 (1965); **186**, 203 (1965).

the diagonal and nondiagonal elements of the density matrix (2.8) in a different way we treat them separately.

(a) Equations for $\dot{\varrho}_{ij}$, $i\neq j$

The spontaneous emission, the nonradiative decay, and the pump destroy the relative phase of the atomic levels i and k after a time called $T_{ik}=\Gamma_{ik}^{-1}$. This effect is described by adding an imaginary part to the real frequencies of the levels. The equation of motion for the nondiagonal elements of the density matrix ρ_{ij} then reads

$$\begin{aligned}\dot{\rho}_{32,\mu}&=i\epsilon_{32}\rho_{32,\mu}-iX_\mu\rho_{31,\mu}+iP_{23,\mu}Y_\mu^\dagger\,,\\ \dot{\rho}_{31,\mu}&=i\epsilon_{31}\rho_{31,\mu}+iY_\mu^\dagger\rho_{21,\mu}-iX_\mu^\dagger\rho_{32,\mu}\,,\\ \dot{\rho}_{21,\mu}&=i\epsilon_{21}\rho_{21,\mu}+iP_{12,\mu}X_\mu^\dagger+iY_\mu\rho_{21,\mu}\,,\end{aligned}\tag{2.9}$$

where

$$\epsilon_{ik}=\epsilon_i-\epsilon_k+i\Gamma_{ik};\quad h_{21,\mu}b_1^\dagger=X_\mu^\dagger;$$
$$h_{23,\mu}b_2^\dagger=Y_\mu^\dagger;\quad P_{ik}=\rho_{ii}-\rho_{kk}\,.$$

(b) Equations for $\dot{\varrho}_{ii}$

We now write down the equations for the diagonal elements of ρ:

$$\begin{aligned}\dot{\rho}_{11,\mu}&=-i\rho_{12,\mu}X_\mu^\dagger+i\rho_{21,\mu}X_\mu+(d_1-\rho_{11,\mu})/T_1+\rho_{22,\mu}/T_1'\,,\\ \dot{\rho}_{22,\mu}&=-i\rho_{23,\mu}Y_\mu^\dagger+i\rho_{32,\mu}Y_\mu+i\rho_{12,\mu}X_\mu^\dagger-i\rho_{21,\mu}X_\mu\\ &\quad+(d_2-\rho_{22,\mu})/T_2+(\rho_{33,\mu})/T_2'\,,\\ \dot{\rho}_{33,\mu}&=i\rho_{23,\mu}Y_\mu^\dagger-i\rho_{32,\mu}Y_\mu+(d_3-\rho_{33,\mu})/T_3\,.\end{aligned}\tag{2.10}$$

In establishing (2.10), the pumping process as well as nonradiative and spontaneous radiative transitions are taken into account. These processes, which manifest themselves in all the expressions in (2.10) containing relaxation times T, are in detail the following:

(α) *The Decay*

Level one decays into the "reservoir," which consists of one or a set of lower levels with a decay time $\tilde{T}_1^{-1}=2\gamma_{10}$. For radiative transitions γ_{10} is the total half-width of level one (the difference between $\tilde{T}_1^{-1}$ and T_1^{-1} etc. will be explained below).

Level two decays both into the "reservoir" and into level one with a decay time $\tilde{T}_2^{-1}=2\gamma_{21}+2\gamma_{20}$ (where for radiative transitions the γ's are the corresponding partial half-width of the homogeneously broadened part of the line). For level three we assume decay into level two and into the reservoir:

$$\tilde{T}_3^{-1}=2\gamma_{32}+2\gamma_{30}\,.$$

We do not consider, however, a direct recombination from 3 to 1, because in the experiments being discussed in our present paper, no allowed optical transition between these states exists nor is there experimental evidence for a nonradiative transition. An example for the different ways of decay is given in Fig. 3.

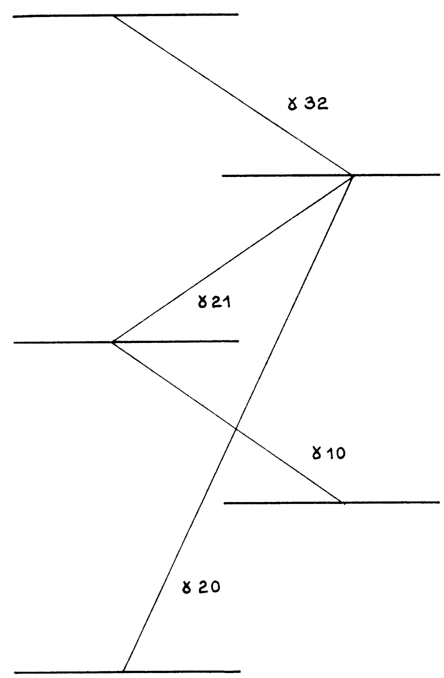

FIG. 3. Schematic diagram of laser cascade. The cascade process occurs between the three upper levels; the two lowest levels serve as reservoir. The γ's represent the radiative half-widths.

(β) *Increase of Occupation Number*

The transitions considered above also give rise, of course, to a corresponding increase of the occupation numbers of the lower levels:

level 2: $\rho_{33,\mu}/T_2'$ (where $T_2'^{-1}=2\gamma_{32}$)

and

level 1: $\rho_{22,\mu}/T_1'$ (where $T_1'^{-1}=2\gamma_{21}$).

(γ) *The Pumping Process*

In the following we wish to derive solutions for the general case, i.e., that in which all three levels participating in the cascade are pumped. A completely exact treatment of this problem would require taking into account explicitly the set of levels composing the "reservoir." In case, the occupation of the cascading levels one, two, three is considerably smaller than that of the reservoir, we may circumvent the above procedure, which would have led to a most lengthy calculation. Under this assumption the pumping of one level can be treated as independent of the fate of the other (laser active) levels, provided the levels are pumped directly from the reservoir. If the pumping alone brings level i after a mean pumping time T_{ip} to an equilibrium occupation number $n_i(0)$, the pumping term reads

$$[n_i(0)-\rho_{ii}]/T_{i,p}\quad\text{for level } i.$$

We now collect all the terms described under (α) and

(γ), which gives for *level three*

$$(n_3(0)-\rho_{33})/T_{3,p}-\rho_{33}/\tilde{T}_3. \tag{2.11}$$

Putting

$$T_{3,p}^{-1}+\tilde{T}_3^{-1}=T_3^{-1} \quad\text{and}\quad n_3(0)/T_{3,p}=d_3/T_3 \tag{2.12}$$

simplifies (2.11) to

$$(d_3-\rho_{33})/T_3. \tag{2.13}$$

In the cases of practical interest $T_{3p}^{-1}<\tilde{T}_3^{-1}$. As follows from (2.12) a decrease in the pumping time or an increase of $n_3(0)$ leads directly to an increase of d_3.

In exactly the same way we introduce

$$T_2^{-1}=T_{2,p}^{-1}+\tilde{T}_2^{-1};\quad n_2(0)/T_{2,p}=d_2/T_2 \tag{2.14}$$

and

$$T_1^{-1}=T_{1,p}^{-1}+\tilde{T}_1^{-1};\quad n_1(0)/T_{1,p}=d_1/T_1. \tag{2.15}$$

These are now finally the quantities occurring in Eqs. (2.10) and which were introduced in order to write (2.10) as concisely as possible.

Finally we have to write down the equations of motion for the light amplitude

$$\begin{aligned}\dot{b}_1^\dagger&=i\omega_1 b_1^\dagger+i\sum_\mu h_{12,\mu}^*\rho_{21,\mu},\\ \dot{b}_2^\dagger&=i\omega_2 b_2^\dagger+i\sum_\mu h_{23,\mu}^*\rho_{32,\mu}.\end{aligned} \tag{2.16}$$

The $b^\dagger$'s are now to be understood as expectation values of the light field, in order to be consistent with the density-matrix formalism as introduced above.[20] ω_λ possesses an imaginary part κ_λ which describes the finite lifetime of mode λ in the cavity: $1/t_\lambda=2\kappa_\lambda$.

In order to solve the coupled equations (2.9), (2.10) and (2.16) we make the following hypothesis:

$$\begin{aligned}&b_\lambda^\dagger=e^{i\Omega_\lambda t}b_\lambda^\dagger(0),\\ &\rho_{32}=e^{i\Omega_2 t}\rho_{32}(0), &&\rho_{11}=\rho_{11}(0)\\ &\rho_{21}=e^{i\Omega_1 t}\rho_{21}(0), &&\rho_{22}=\rho_{22}(0)\\ &\rho_{31}=e^{i(\Omega_1+\Omega_2)t}\rho_{31}(0), &&\rho_{33}=\rho_{33}(0).\end{aligned} \tag{2.17}$$

It can be very easily checked that the whole time dependence drops out of the equations, which are now purely algebraic in nature. From the set of Eqs. (2.9) we can now immediately eliminate the off-diagonal elements for which we obtain the following expressions:

$$\begin{aligned}\rho_{32,\mu}&=(1/D_\mu)Y_\mu^\dagger[P_{23,\mu}(\Delta_{31}\Delta_{21}-\tilde{n}_{2,\mu})-P_{12,\mu}\tilde{n}_{1,\mu}],\\ \rho_{21,\mu}&=(1/D_\mu)X_\mu^\dagger[P_{12,\mu}(\Delta_{32}\Delta_{31}-\tilde{n}_{1,\mu})-P_{23,\mu}\tilde{n}_{2,\mu}],\\ \rho_{31,\mu}&=(1/D_\mu)X_\mu^\dagger Y_\mu^\dagger(P_{12,\mu}\Delta_{32}-P_{23,\mu}\Delta_{21}),\end{aligned} \tag{2.18}$$

where

$$\begin{aligned}\Delta_{ik}&=\Omega_{ik}-(\epsilon_i-\epsilon_k)-i\Gamma_{ik};\\ &\Omega_{ik}=\Omega_1\quad\text{for}\quad i=2,\ k=1\\ &\qquad=\Omega_2\quad\text{for}\quad i=3,\ k=2\\ D_\mu&=\Delta_{32}\Delta_{31}\Delta_{21}-\tilde{n}_{1,\mu}\Delta_{21}-\tilde{n}_{2,\mu}\Delta_{32},\\ \tilde{n}_{1,\mu}&=|X_\mu|^2;\quad \tilde{n}_{2,\mu}=|Y_\mu|^2.\end{aligned} \tag{2.19}$$

3. RATE EQUATIONS AND Γ_{31} APPROXIMATION

As can be shown by detailed calculation the results are not altered significantly if the phase-memory time between level 1 and 3 is taken to be very short. In this case, the above expressions can be simplified as follows:

$$\begin{aligned}\rho_{32,\mu}&=P_{23,\mu}Y_\mu^\dagger/\Delta_{32},\\ \rho_{21,\mu}&=P_{12,\mu}X_\mu^\dagger/\Delta_{21},\\ \rho_{31,\mu}&=0.\end{aligned} \tag{3.1}$$

Inserting the expressions for the off-diagonal elements into the equations for the diagonal elements and into the equations of motion for the light amplitudes we obtain:

$$\begin{aligned}0&=-P_{12,\mu}n_1W_{21,\mu}+(d_1-\rho_{11,\mu})/T_1+\rho_{22,\mu}/T_1',\\ 0&=-P_{23,\mu}n_2W_{32,\mu}+P_{12,\mu}n_1W_{21,\mu}\\ &\qquad+(d_2-\rho_{22,\mu})/T_2+\rho_{33,\mu}/T_2',\\ 0&=P_{23,\mu}n_2W_{32,\mu}+(d_3-\rho_{32,\mu})/T_3,\end{aligned} \tag{3.2}$$

and

$$\begin{aligned}i\Omega_1 b_1^\dagger&=i\omega_1 b_1^\dagger+i\{\sum_\mu|h_{12,\mu}|^2(P_{12,\mu}/\Delta_{21})\}b_1^\dagger,\\ i\Omega_2 b_2^\dagger&=i\omega_2 b_2^\dagger+i\{\sum_\mu|h_{23,\mu}|^2(P_{23,\mu}/\Delta_{32})\}b_2^\dagger,\end{aligned} \tag{3.3}$$

where we have used the following abbreviations:

$$W_{ik,\mu}=\frac{2\Gamma_{ik}}{[\Omega_{ik}-(\epsilon_i-\epsilon_k)]^2+\Gamma_{ik}^2}\times|h_{ik,\mu}|^2;$$

$$n_\lambda \text{ number of photons}. \tag{3.3a}$$

If we identify the diagonal elements of the density matrix $\rho_{ii,\mu}$ with the occupation number $N_{i,\mu}$ of energy level i at atom μ we immediately find the following equations

$$\begin{aligned}dN_{1,\mu}/dt&=0=(N_{2,\mu}-N_{1,\mu})n_1W_{21,\mu}\\ &\qquad+(d_1-N_{1,\mu})/T_1+N_{2,\mu}/T_1',\\ dN_{2,\mu}/dt&=0=(N_{3,\mu}-N_{2,\mu})n_2W_{32,\mu}\\ &\qquad+(N_{1,\mu}-N_{2,\mu})n_1W_{21,\mu}\\ &\qquad+(d_2-N_{2,\mu})/T_2+N_{3,\mu}/T_2',\\ dN_{3,\mu}/dt&=0=(N_{2,\mu}-N_{3,\mu})n_2W_{32,\mu}+(d_3-N_{3,\mu})/T_3,\end{aligned} \tag{3.4}$$

which are in fact equivalent to the well-known rate equations for the electronic occupation numbers.

After division of (3.3) by $b_1^\dagger$ and $b_2^\dagger$, respectively, we obtain for the real part

$$\begin{aligned}2\kappa_1&=\sum_\mu W_{21,\mu}(N_{2,\mu}-N_{1,\mu}),\\ 2\kappa_2&=\sum_\mu W_{32,\mu}(N_{3,\mu}-N_{2,\mu}),\end{aligned} \tag{3.5}$$

where κ_i is inversely proportional to the lifetime of mode i. Therefore in the large-Γ_{31} approximation the whole problem is reduced to the usual rate equations.

Our treatment, however, immediately yields the frequency shifts in case of a homogeneously broadened line.[21] If we take the imaginary part of Eq. (3.3) we find the following expression for the frequencies Ω_λ:

$$\Omega_1=\frac{\Gamma_{21}\omega_1+\kappa_1(\epsilon_2-\epsilon_1)}{\Gamma_{21}+\kappa_1};\quad \Omega_2=\frac{\Gamma_{32}\omega_2+\kappa_2(\epsilon_3-\epsilon_2)}{\Gamma_{32}+\kappa_2}. \quad (3.6)$$

4. SOLID-STATE LASER WITH A HOMOGENEOUSLY BROADENED LINE

From Eq. (3.4) the occupation numbers of the electronic levels can be easily determined as a function of the photon numbers. Because we need only the differences between the photon numbers, we write down the corresponding expressions:

$$N_{2,\mu}-N_{1,\mu}=D_N^{-1}[-(d_1/T_1)((T_2T_3)^{-1}+W_{32,\mu}n_2(T_2^{-1}+T_3^{-1}-T_2'^{-1}))+(d_2/T_2)((T_1T_3)^{-1}-(T_3T_1')^{-1}$$
$$+W_{32,\mu}n_2(T_1^{-1}-T_1'^{-1}))+(d_3/T_3)((T_1T_2')^{-1}-(T_1'T_2')^{-1}+W_{32,\mu}n_2(T_1^{-1}-T_1'^{-1}))], \quad (4.1)$$

and

$$N_{3,\mu}-N_{2,\mu}=D_N^{-1}[-(d_1/T_1T_3)W_{21,\mu}n_1-(d_2/T_2)((T_1T_3)^{-1}+W_{21,\mu}n_1(T_3)^{-1})$$
$$+(d_3/T_3)((T_1T_2)^{-1}(1-T_2/T_2')+W_{21,\mu}n_1(T_1^{-1}+T_2^{-1}-T_1'^{-1}-T_2'^{-1}))], \quad (4.2)$$

where

$$D_N=(T_1T_2T_3)^{-1}+W_{21,\mu}n_1T_3^{-1}(T_2^{-1}+T_1^{-1}-T_1'^{-1})+W_{32,\mu}n_2T_1^{-1}(T_2^{-1}+T_3^{-1}-T_2'^{-1})$$
$$+W_{21,\mu}n_1W_{32,\mu}n_2(T_1^{-1}+T_2^{-1}+T_3^{-1}-T_1'^{-1}-T_2'^{-1}). \quad (4.3)$$

In the final step of our analysis we have to insert the expressions (4.1) and (4.2) into (3.5) and to perform the sum over the atomic positions. As is known from other examples this summation (or integration) represents essential difficulties if the exact expressions for N_2-N_1 or N_3-N_1 are used. Fortunately, however, the expressions containing the n's are small enough, so that one can expand N_2-N_1 and N_3-N_2 into power series, where only expressions up to and including the second power in the n_i are to be considered for the cascade effect.

The equation for mode one then reads

$$\kappa_1=\frac{\Gamma_{21}}{(\Delta\Omega_1)^2+\Gamma_{21}^2}\{\cdots\}, \quad (4.4)$$

where

$$\{\cdots\}=g_1^2\rho A+(n_1/V)\rho\tilde{W}_{21}(\tfrac{3}{2})^3g_1^4B+(n_2/V)g_1^2g_2^2\rho\tilde{W}_{32}C$$
$$+(n_1/V)(n_2/V)g_1^4g_2^2\tilde{W}_{21}\tilde{W}_{32}\rho(\tfrac{3}{2})^3D+g_1^2g_2^4\tilde{W}_{32}^2(n_2/V)^2(\tfrac{3}{2})^3\rho E, \quad (4.5)$$

where

$$\tilde{W}_{21}=W_{21,\mu}/|h_{21,\mu}|^2;\quad \tilde{W}_{32}=W_{32,\mu}/|h_{32,\mu}|^2. \quad (4.6)$$

We have used the abbreviation $g_1=|G_{21}|$ and $g_2=|G_{32}|$ for the G's introduced in (2.5). The W's are defined in (3.3a) and

$$\begin{aligned}
A&=[-d_1+d_2(1-T_1/T_1')+d_3(T_2/T_2'-T_1T_2/T_1'T_2')],\\
B&=(T_1+T_2-T_2T_1/T_1')[d_1-d_2(1-T_1/T_1')-d_3(T_2/T_2')(1-T_1/T_1')],\\
C&=T_2(1-T_1/T_1')(1-T_3/T_2')[-d_2+d_3(1-T_2/T_2')],\\
D&=(1-T_1/T_1')(1-T_3/T_2')\{-d_1T_2^2+d_2[T_1T_2+2T_2^2(1-T_1/T_1')]\\
&\qquad -d_3[T_2^2(1-T_1/T_1')+T_1T_2(1-T_2/T_2')-2(T_2^3/T_2')(1-T_1/T_1')]\},\\
E&=(1-T_1/T_1')(T_2+T_3-T_2T_3/T_2')\{d_2T_2(1-T_3/T_2')+d_3T_2[(1/T_2')(T_2+T_3-T_2T_3/T_2')-1]\}.
\end{aligned} \quad (4.7)$$

ρ is the density of the atoms. The factor $(\frac{3}{2})^3$ stems from the assumption that the modes of the two light waves differ not only in axial direction, but also in radial direction. If the modes differ only in axial direction, but have the same angular and radial dependence, this factor is to be replaced by $\frac{3}{2}$. As has been shown in a previous paper[22] these factors give rise to the coexistence of modes of a solid-state laser within a linewidth of one transition, even if the line is homogeneously broadened.[23] This mechanism may also apply at present so that we expect that if the lower transition oscillates, then the upper one is pumped so highly that several modes oscillate simultaneously. In this case our results for the laser cascade can be generalized simply by taking a suitable average over the upper modes and n_2 now stands for the average number of photons of these modes. In gas lasers, on the other hand, this factor $(\frac{3}{2})$ does not occur (in lowest approximation) due to the

[21] C. H. Townes, *Second Quantum Electronics Conference*, edited by R. J. Singer (Columbia University Press, New York, 1961), p. 3.

[22] H. Haken and H. Sauermann, Z. Physik **173**, 261 (1963).

[23] A similar result has been published by C. L. Tang, H. Statz, and G. A. de Mars, J. Appl. Phys. **34**, 2289 (1963).

motion of atoms. In fact, the coexistence of modes is accomplished in gas lasers by the inhomogeneously broadened line due to Doppler broadening.

The laser condition for mode 1 can be found by putting $n_1=0$ which yields

$$\kappa_1=\frac{\Gamma_{21}}{(\Delta\Omega_1)^2+\Gamma_{21}^2}\left\{g_1^2\rho A+\left(\frac{n_2}{V}\right)g_1^2g_2^2\rho\tilde{W}_{32}C+g_1^2g_2^4\tilde{W}_{32}\left(\frac{n_2}{V}\right)^2(\tfrac{3}{2})^3\rho E\right\}. \quad (4.8)$$

In it the quantities A, C and E are defined above. For mode two it is sufficient to derive the expressions up to linear terms in n_1 and n_2. We then have

$$\kappa_2=[\Gamma_{32}/((\Delta\Omega_2)^2+\Gamma_{32}^2)]\{\cdots\}, \quad (4.9)$$

where

$$\{\cdots\}=g_2^2\rho A'+(n_1/V)\rho\tilde{W}_{21}g_1^2g_2^2B'+(n_2/V)\rho\tilde{W}_{32}C'g_2^4(\tfrac{3}{2})^3, \quad (4.10)$$

and where

$$\begin{aligned}A'&=-d_2+d_3(1-T_2/T_2'),\\ B'&=-d_1T_2+d_2T_2(1-T_1/T_1')+d_3T_2^2(T_2')^{-1}(1-T_1/T_1'), \quad (4.11)\\ C'&=d_2(T_2+T_3-T_3(T_2/T_2'))-d_3(1-T_2/T_2')(T_2+T_3-T_3(T_2/T_2')).\end{aligned}$$

5. SOLID-STATE LASER WITH AN INHOMOGENEOUSLY BROADENED LINE

We assume now that there is a spread of transition frequencies ϵ_2-ϵ_1 and ϵ_3-ϵ_2. This requires summing up in Eqs. (3.5) and (4.4), respectively, not only over the atomic position but also taking a weighted average over the spread of frequencies. To this end we assume independent Gaussian distributions for ϵ_2-ϵ_1 and ϵ_3-ϵ_2. Instead of Eq. (4.4) for mode 1 we now obtain

$$\kappa_1=\int\int\frac{d\epsilon_{21}d\epsilon_{32}}{\pi\alpha_1\alpha_2}\exp\left[-\left(\frac{\epsilon_{21}-\epsilon_{21}^0}{\alpha_1}\right)^2\right]\exp\left[-\left(\frac{\epsilon_{32}-\epsilon_{32}^0}{\alpha_2}\right)^2\right]\frac{\Gamma_{21}}{(\Omega_1-\epsilon_{21})^2+\Gamma_{21}^2}\{\cdots\}, \quad (5.1)$$

where the bracket $\{\cdots\}$ is the same as in Eq. (4.4). The same type of integrals which occur here have been considered in a previous paper.[15] One thus obtains readily

$$\begin{aligned}\kappa_1=&g_1^2\rho A\frac{\pi^{1/2}}{\alpha_1}\exp(-\delta_1^2)+\left(\frac{n_1}{V}\right)\rho(\tfrac{3}{2})^3g_1^4B\pi^{1/2}\exp(-\delta_1^2)/\Gamma_{21}\alpha_1+\left(\frac{n_2}{V}\right)g_1^2g_2^2\rho C\frac{2\pi}{\alpha_1\alpha_2}\exp(-\delta_1^2)\exp(-\delta_2^2)\\ &+\left(\frac{n_1}{V}\right)\left(\frac{n_2}{V}\right)g_1^4g_2^2\rho(\tfrac{3}{2})^3D2\pi\frac{\exp[-(\delta_1^2+\delta_2^2)]}{\alpha_1\alpha_2\Gamma_{21}}+\left(\frac{n_2}{V}\right)^2g_1^2g_2^4\rho E\frac{2\pi}{\Gamma_{32}\alpha_1\alpha_2}\exp[-(\delta_1^2+\delta_2^2)]. \quad (5.2)\end{aligned}$$

The quantities A, B, C, D, E are the same as defined in (4.7);

δ_1 is given by $(\Omega_1-\epsilon_{21}^0)/\alpha_1$, δ_2 by $(\Omega_2-\epsilon_{32}^0)/\alpha_2$.

If we consider optical transitions close to resonance, we may replace the exponential factors by 1. As one may note, the former factor Γ_{21}^{-1} (for resonance) in front of the bracket in (4.4) is now replaced by $\pi^{1/2}/\alpha_1$. Different factors occur, however, for the n's.

In a completely analogous manner we obtain for mode 2:

$$\begin{aligned}\kappa_2=&g_2^2\rho A'(\pi^{1/2}/\alpha_2)\exp(-\delta_2^2)\\ &+(n_1/V)\rho g_1^2g_2^2B'(2\pi/\alpha_1\alpha_2)\exp[-(\delta_1^2+\delta_2^2)]\\ &+(n_2/V)\rho C'g_2^4(\tfrac{3}{2})^3(\pi^{1/2}/\Gamma_{32}\alpha_2)\exp(-\delta_2^2), \quad (5.3)\end{aligned}$$

where A', B' and C' are defined in (4.11).

6. DISCUSSION OF RESULTS

Because the coefficients of the n's occurring in Eqs. (5.2) and (5.3) are rather complicated and since there are cascades in which only the upper level is pumped from the reservoir, let us consider this case in more detail. With $d_1=d_2=0$ the constants of Eq. (5.2) reduce to

$$\begin{aligned}A&=(T_2/T_2')(1-T_1/T_1')d_3,\\ B&=-d_3(T_1+T_2-T_1T_2/T_1')(T_2/T_2')(1-T_1/T_1'),\\ C&=T_2(1-T_1/T_1')(1-T_3/T_2')(1-T_2/T_2')d_3,\\ E&=(1-T_1/T_1')(T_2+T_3-T_2T_3/T_2')\times[(1/T_2')(T_2+T_3-T_2T_3/T_2')-1]d_3. \quad (6.1)\end{aligned}$$

(D is not important for what follows.) As is immediately seen, all relaxation times which were introduced into the equations for the diagonal elements (2.10) occur. According to the definitions following Eq. (2.10) we have for the present case (only level three pumped)

$$\begin{aligned}&T_{p1}^{-1}=T_{p2}^{-1}=0;\\ &T_1^{-1}=2\gamma_{10},\quad T_1'^{-1}=2\gamma_{21},\quad T_2^{-1}=2\gamma_{21}+2\gamma_{20}, \quad (6.2)\\ &T_2'^{-1}=2\gamma_{32};\quad T_3'^{-1}=T_{3p}^{-1}+2\gamma_{32}+2\gamma_{30}.\end{aligned}$$

For the following discussion we assume that the γ_{ik}'s are the radiative half-widths of the atomic lines corresponding to the spontaneous decay from the state i to the state k. If $k=0$, γ_{ik} will mean the sum of the widths corresponding to transitions to a set of "ground levels." For a complete discussion we have to know γ_{10}, γ_{21}, γ_{20}, γ_{32}, γ_{30}, and T_{3p}. It should be noted that γ_{30} occurs only jointly with T_{3p}^{-1} so that we introduce $T_{3p}^{-1}+2\gamma_{30}\equiv T_{3p}'^{-1}$. In the He-Ne gas laser one has $T_{3p}^{-1}<2\gamma_{32}$.

If we choose *as an explicit example the pure neon cascade* described above, the following γ's are known experimentally:

$$\gamma_{21}\simeq 10^7\ \text{sec}^{-1};^{24}\quad \gamma_{10}\simeq 10^8\ \text{sec}^{-1}.^{25}$$

In the following we neglect γ_{20} as compared to γ_{21}. This is justified because the lifetime τ_{20} of the $2s_2$ state with respect to a decay to the ground state is appreciably increased due to light-trapping (in the experimental pressure range of 10^{-1}-10^{-2} Torr depending on the tube diameter). On the other hand γ_{32} is not well known, whereas the total half-width $\gamma_{32}+\gamma_{30}$ has been recently measured by Klose, who finds $(\gamma_{32}+\gamma_{30})^{-1}=63.5$ nsec,[26] so that $\gamma_{32}+\gamma_{30}$ is about 1, 5 times larger than γ_{21}. Thus we have an order-of-magnitude estimate of γ_{32}. As we will see below, however, the occurrence of a laser cascade depends on whether $\gamma_{32}>\gamma_{21}$ or $\gamma_{32}<\gamma_{21}$. We can now write down the coefficients A, C, E, which occur in the threshold condition of the lower mode both for a homogeneously and an inhomogeneously broadened line:

$$\begin{aligned}
A&=(\gamma_{32}/\gamma_{21})(1-\gamma_{21}/\gamma_{10})d_3\,,\\
B&=-d_3(\gamma_{32}/2\gamma_{21}^2)(1-\gamma_{21}/\gamma_{10})\,,\\
C&=(2\gamma_{21})^{-1}(1-\gamma_{21}/\gamma_{10})(1+2\gamma_{32}T_{3p}')^{-1}(1-\gamma_{32}/\gamma_{21})d_3\,,\\
E&=\left(1-\frac{\gamma_{21}}{\gamma_{10}}\right)\frac{1+2T_{3p}'\gamma_{21}}{(1+2\gamma_{32}T_{3p}')^2}\\
&\qquad\times(4\gamma_{21}^2)^{-1}(1-\gamma_{32}/\gamma_{21})(-d_3)\,,\\
A'&=(1-\gamma_{32}/\gamma_{21})d_3\,,\\
B'&=(\gamma_{32}/2\gamma_{21}^2)(1-\gamma_{21}/\gamma_{10})d_3\,,\\
C'&=-d_3\left(1-\frac{\gamma_{32}}{\gamma_{21}}\right)\frac{1+2T_{3p}'\gamma_{21}}{1+2T_{3p}'\gamma_{32}}\cdot(2\gamma_{21})^{-1}.
\end{aligned}\tag{6.3}$$

Since $\gamma_{21}<\gamma_{10}$, one has A, $B'>0$, $B<0$. The sign of C and E still depends on the relative size of γ_{32} and γ_{21}. Let us assume first, that $\gamma_{32}>\gamma_{21}$. Then $C<0$, $A'<0$, $E>0$ and $C'>0$. Equation (5.3) can be put into the form

$$\kappa_2/d_3+\alpha'=\beta' n_1+\gamma' n_2\,,\tag{6.4}$$

where α', β', $\gamma'>0$ and do not depend on the pumping occupation d_3. This formula shows that in this case, where $\gamma_{32}>\gamma_{21}$, no cascade can occur. If the inversion $\sim d_3$ increases, the total output of either n_1 or n_2 or both must decrease.

Let us consider the opposite case: $\gamma_{32}<\gamma_{21}$. Then $C>0$, $A'>0$, $E<0$ and $C'<0$. Equation (5.3) then has the form

$$\alpha'-\kappa_2/d_3=\gamma' n_2-\beta' n_1\,,\tag{6.5}$$

where again α', β', $\gamma'>0$. For not too high an inversion the photon numbers are small, so that we may drop in the laser Eq. (5.2) for mode-one terms which are bilinear or quadratic in n_1, n_2. The laser equation for mode one then takes the form

$$\alpha-\kappa_1/d_3=n_1\beta-\gamma n_2\,,\tag{6.6}$$

with α, β, $\gamma>0$.

Equations (6.5) and (6.6) can be solved consistently only if $n_1>0$ and $n_2>0$. Consider for example the case where we approach $n_1=0$ by lowering the pumping. This implies that $b_1^\dagger=0$. In deriving (3.5) from (3.3) we divided both sides of (3.3) by $b_1^\dagger$. Therefore (6.6), which follows from (3.5), is no longer valid and has to be dropped. The same reasoning applies, *mutatis mutandis*, to n_2. This line can be continued also when $n_i<0$ because the photon number cannot be negative. This procedure is exactly the same as the one used to demonstrate the possible coexistence of modes in a homo-

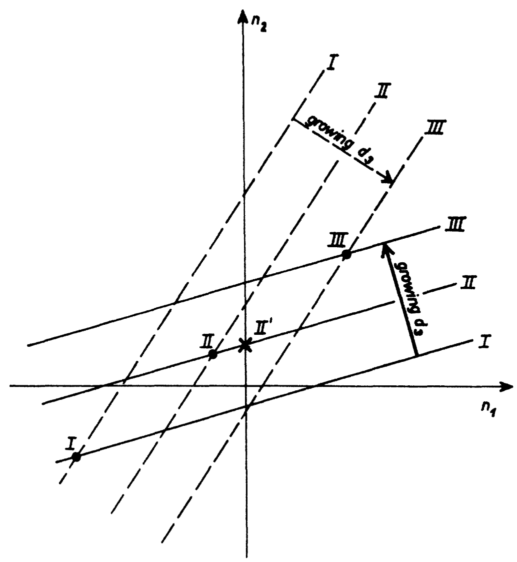

FIG. 4. Schematic plot of n_2 versus n_1. The dashed lines represent Eq. (6.6), the solid lines Eq. (6.4), for different values of the inversion parameter d_3. The points I, II, III represent situations where no mode shows laser action, mode 2 shows laser action, and both modes show laser action in a cascade, respectively.

[24] W. R. Bennett, Jr., *Second Quantum Electronics Conference*, edited by J. R. Singer (Columbia University Press, New York, 1961), p. 28.

[25] R. Ladenburg, Rev. Mod. Phys. 5, 243 (1933).

[26] J. Z. Klose, Bull. Am. Phys. Soc. 9, 425 (1964). One of the authors (R. A.) is indebted to Dr. Klose for communication before publication of his lifetime measurements concerning some $3p$ levels of neon.

geneously broadened line.[22,23] A deeper foundation of this procedure has been indicated recently.[27]

We shall now discuss Eqs. (6.5) and (6.6) by a graphical plot (Fig. 4). Figure 4 demonstrates three typical situations, according to three different pump rates. The dashed lines represent Eq. (6.6), the solid lines Eq. (6.5). For low pumping we obtain the lines I. Their intersection lies in the third quadrant, i.e., both n's are negative and no laser cascade occurs. Therefore we check, if single-mode action may occur, by dropping one of the Eqs. (6.5), (6.6). In the representation of Fig. 4 this means, that we have to see if the dashed line allows for $n_1>0$, $n_2=0$ or the solid line for $n_2>0$, $n_1=0$, which is, in the present example, not the case. Therefore no laser action occurs at all. Consider now the lines II, where a somewhat increased pumping rate is assumed. The intersection still implies $n_1<0$ and no cascade can occur. However, the solid line cuts the n_2 axis at a positive value (II′) which indicates that we are now (slightly) above threshold for the upper laser transition. For still higher pumping we obtain finally the lines III, having a crossing point for $n_1>0$, $n_2>0$. Here the full cascade occurs. Figure 4 allows a condition for the occurrence of a cascade to be deduced. The crossing point shifts with increasing d_3 (pumping) from the third to the first quadrant only, if the slope of the dashed lines is steeper than that of the solid lines or, if

$$\beta/\beta'>\gamma/\gamma'. \tag{6.7}$$

For the cascade described in the beginning of and treated throughout this paragraph, condition (6.7) reads, after insertion of the explicit expressions for β, β', γ, γ'

$$(\tfrac{3}{2})^6(1+2T_{3p}'\gamma_{21})\frac{\alpha_1\alpha_2}{\Gamma_{21}\Gamma_{32}2\pi}>\left(1-\frac{\gamma_{21}}{\gamma_{10}}\right)$$

(inhomogeneously broadened line) (6.8)

which is automatically fulfilled since $\alpha_1>\Gamma_{21}$ and $\alpha_2>\Gamma_{32}$. For comparison we quote the explicit form of (6.7) for a homogeneously broadened line:

$$(\tfrac{3}{2})^6(1+2T_{3p}'\gamma_{21})>(1-\gamma_{21}/\gamma_{10}), \tag{6.9}$$

which is again always fulfilled.

It is a remarkable feature of Eqs. (6.5) and (6.6) that n_1 and n_2 play a completely symmetrical role. Thus, not only laser action of the upper transition can cause laser action of the lower one, but also the inverse can happen. Therefore the question arises, which of both transitions occurs first and may, at higher pumping, trigger the other transition. Inspection of Eqs. (6.5), (6.6) shows, that the line which lases first is the one, for which the corresponding Eq. (6.5) or (6.6) has an inhomogeneous term which is bigger than that of the remaining equation. Let us assume, that we have, as usual, to deal with longitudinal modes, for which the losses are equal $\kappa_1=\kappa_2$. We thus are left to discuss α/α'.

Provided the upper and lower transition have the same inhomogeneous broadening (which is, of course, the case for Doppler-broadening), we find by insertion of the expressions for α and α':

$$\alpha/\alpha'=(g_1^2/g_2^2)(\gamma_{32}/\gamma_{21})(1-\gamma_{21}/\gamma_{10})/(1-\gamma_{32}/\gamma_{21}). \tag{6.10}$$

Because the radiative half-width γ is proportional to the absolute square of the optical matrix element g^2 and to the density of light-field states at that specific frequency $\bar{\rho}(\omega)$, i.e., $\gamma\sim g^2\rho$ (6.10) can be reduced to

$$\alpha/\alpha'=\bar{\rho}(\omega_2)/\bar{\rho}(\omega_1)(1-\gamma_{21}/\gamma_{10})/(1-\gamma_{32}/\gamma_{21}). \tag{6.11}$$

For the two-step cascade in pure neon, as described in the introduction and considered in this chapter, we have

$$\alpha/\alpha'\approx\tfrac{1}{4}(1-\gamma_{32}/\gamma_{21})^{-1}. \tag{6.12}$$

Experimentally it is found, that with wide-band mirrors the upper transition starts laser action first, so that we conclude $\alpha<\alpha'$ (6.13) or, from (6.12) $\gamma_{32}<\frac{3}{4}\gamma_{21}$ (6.14) which puts a new limit on γ_{32}. Once the assignment (6.14) is made Eqs. (6.5) and (6.6) also readily describe the following experimental findings:

If one takes dielectric (narrow-band) mirrors, two cases can be distinguished:

(1) If the maximum reflectivity of the mirrors is centered around the upper transition we observe that the gain of the upper transition is not changed as compared to the case of wide-band mirrors. Laser action occurs only in the upper transition.

(2) If the maximum reflectivity is centered around the lower transition, the gain of the lower transition is reduced compared to the wide-band mirror case.[12]

It is obvious that the treatment outlined above can also be applied to the general case of an arbitrary pumping of all three levels, thus allowing to check quickly, if, in a given system, a laser cascade can occur or not.

We conclude with a general remark about the question of whether, in the cascading process, phase information is transferred from the upper to the lower mode. By this we understand the following: As has been derived theoretically,[27,28] and is now also well established experimentally[29,30] the complex amplitude of a laser-light mode above threshold can be written in the form $\exp[i\omega t+i\varphi(t)](r_0+s)$ where $\varphi(t)$ is the fluctuating phase, r_0 a stable (real) amplitude and s represents small amplitude fluctuations. For the laser cascade one may ask if a systematic phase modulation by external means or also noisy phase fluctuations of the mode belonging to the upper (lower) transition causes changes in the phase of that belonging to the lower (upper) transition. This question can be answered in the follow-

[27] H. Haken, Z. Physik **181**, 96 (1964); **182**, 346 (1965).

[28] H. Haken, Phys. Rev. Letters **13**, 329 (1964).

[29] J. A. Armstrong and A. W. Smith, Phys. Rev. Letters **14**, 68 (1965).

[30] Ch. Freed and H. A. Haus, Appl. Phys. Letters **6**, 85 (1965).

ing way: As can be seen from (2.9), (2.10) and (2.16) the (complex) *light amplitude* can be *eliminated* from these equations which are still exact within the framework of a density matrix treatment (neglecting antiresonant terms). Therefore only the photon numbers (or light *intensities*) are coupled to each other. This result is well known from other problems, for instance spontaneous Raman scattering: using an intermediate level under resonance conditions destroys the conservation of phase.

It follows that a phase modulation of the lower transition by the upper one (or vice versa) via a cascade is not possible. Equivalently the linewidth of the lower or upper transition should be the same, whether or not the other complementary mode lases (provided the net gain for the single mode remains the same). On the other hand, the amplitude fluctuations s_1, s_2 influence each other. The detailed results of this analysis will be published elsewhere.

PHYSICAL REVIEW VOLUME 140, NUMBER 2A 18 OCTOBER 1965

Electronic g Factors of Low-Lying Levels of Fe I, Cr I, and Mn I†

WILLIAM J. CHILDS AND LEONARD S. GOODMAN
Argonne National Laboratory, Argonne, Illinois
(Received 24 May 1965)

The atomic-beam magnetic-resonance technique was used to measure the electronic g factors of 11 low-lying levels in Fe I, Cr I, and Mn I. Both odd-A isotopes (which show hfs) and even-A isotopes were used for the study. The theoretical implications of the results are discussed.

INTRODUCTION

THE measurement of electronic g factors of the ground state and excited atomic levels is important for both theoretical and practical reasons. The values obtained, if of sufficiently high precision, give immediate and quantitative evidence of the presence of any admixture in the atomic wave functions, and constitute a useful guide to theoretical investigation of the states and their interactions. In addition, knowledge of the g factor of a particular level is of great value to the experimentalist making hfs studies.

In this paper, we report g values for most $J\neq 0$ levels below 11 000 cm^{-1} in Fe I, Cr I, and Mn I. No measurement was made for the $^5F_1$ level in Fe at 8155 cm^{-1}, for which $g_J\approx 0$, or for the $^5S_2$ level in Cr at 7593 cm^{-1}, which could not be distinguished from the much more intense $^7S_3$ ground state. In addition, the four $J\neq 0$ states of the ^5D term in Cr at ~ 8000 cm^{-1} were not resolved into separate components.

PROCEDURE

The experiment was conducted with a conventional atomic-beam magnetic-resonance apparatus. Since the technique is now classic,[1] no description of the general principles will be given here.

The particular apparatus used has been described in detail by Childs *et al.*[1] Ovens of ZrO were used for the Fe and Cr measurements, and graphite ovens were employed for the Mn. The intensity of the homogeneous magnetic field in which the radiofrequency transitions were induced was measured by observing resonances in an independent beam of K^{39}, in which the $(F, m_F \leftrightarrow F', m_F') = (2, -1 \leftrightarrow 2, -2)$ transition was used. The atomic beam was detected with an electron-bombardment universal detector incorporating a mass spectrometer. Details of the add-subtract scaling technique used have been published by Childs *et al.*[1] Many of the observations would not have been possible without such a system.

Fe

The most abundant isotope in the Fe beam was the even-even isotope Fe^{56} (92%). For such an atom with $I=0$, one resonance may be observed for each $J\neq 0$ state of the atom that (1) lives long enough to traverse the apparatus, (2) is sufficiently populated in the atomic beam, and (3) has a large enough electronic gyromagnetic ratio g_J. The transition observed in the state J is that between the magnetic substates with $m_J=+1$ and $m_J=-1$, and its relative intensity is therefore given by the Boltzmann factor.[2] The resonance frequency ν of this double-quantum transition is proportional to the magnetic field H if the field is not too strong. Thus,

$$\nu_J = [E(m_J=+1) - E(m_J=-1)]/2h = g_J\mu_0 H/h\,,$$

where μ_0 is the Bohr magneton and h is Planck's con-

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

[1] J. R. Zacharias, Phys. Rev. **61**, 270 (1942); W. J. Childs, L. S. Goodman, and D. von Ehrenstein, *ibid.* **132**, 2128 (1963).

[2] The relative intensities may not be given simply by the Boltzmann factors if higher order multiple-quantum transitions of the type $(J, m_J>1 \leftrightarrow J, -m_J)$ are induced by overpowering the line.