The actual species present in the equilibrium mixture might be different from the species used for illustration. Perhaps the high-temperature reaction mixtures of the system under study includes species which have not as yet been identified. An oxygen-rich species resultant from interaction of aluminum oxide with hydrogen chloride, more volatile than an oxygen-deficient species, could explain the transport of α-aluminum oxide and the formation of an oxygen-deficient oxide.

The observations of Diamond[4] and Arghiropoulos[5] did not require the presence of hydrogen chloride. Aluminum oxide may be considered as a high molecular weight inorganic polymer made up of condensed Al_2O_3 monomers. The properties, such as vapor pressure, which have been recorded for this material are the properties of the polymer and may be different from the properties expected from a monomer, dimer, or other lower molecular weight species. Hydrogen chloride may be considered as a gaseous catalyst which breaks the bonds between aluminum and oxygen atoms.

Stoichiometrically, there are one and one half oxygen atoms for every aluminum atom. Statistically, as a result of bond breaking, an aluminum atom could end up being bonded to none, one, two, or three oxygen atoms. The final species, in a system where there was complete mobility of atoms, electrons, and chemical bonds, would be the monomer Al_2O_3. At some stage, disproportionation or oxidation–reduction could occur as shown by the following equations:

$$2\ Al_2O_3 \rightarrow Al_2O_2 + Al_2O_4, \tag{4}$$

$$Al_2O_3 \rightarrow AlO + AlO_2. \tag{5}$$

AlO and Al_2O_2 are the monomeric and dimeric forms of aluminum monoxide. An oxygen-rich species resultant from bond breaking, more volatile than the oxygen-deficient species, could explain the transport and deposition of α-aluminum oxide and the formation of a suboxide.

One might consider the black oxide to be a composite of Al_2O_2 and Al_2O_3 molecules. Electron transfer could then occur by an oxidation–reduction process. The material would be expected to have maximum conductivity when the mole fractions were equal, decreasing to the conductivity of the reduced or oxidized oxide as the percentage composition was changed.

To summarize, these results show a new way to transport and deposit aluminum oxide and to make a black oxide. Other methods of aluminum oxide growth require higher temperatures or starting materials that are more difficult to obtain. The black oxide has some properties similar to the oxide observed by Arghiropoulos[5]; i.e., it behaves as a semiconductor and turns white when heated in air. It differs in that metallic aluminum does not vaporize at high temperatures in vacuum, and it is inert in hydrochloric acid. It is possible that the properties of the black oxide, like the properties of the normal white oxide, are dependent on the temperature to which the oxide has been fired. The thesis might be advanced that the higher the temperature to which the oxide has been fired, the higher the molecular weight of the resultant aggregate, and the more inert the substance. The black oxide may be similar to that observed by Diamond; if so, it was formed at a lower temperature.

These observations may be important to (1) the metallurgy of aluminum, (2) the fabrication of activated alumina, (3) the fabrication of aluminum oxide diodes, conductors, and resistors, (4) the sintering of aluminum oxide, and (5) the growth of aluminum oxide.

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Super Radiant Narrowing in Fluorescence Radiation of Inverted Populations

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THE effect of resonance reabsorption of fluorescent radiation on the spectral distribution has been considered as early as 1913 by Landenburg and Reiche.[1] Schein[2] and Zemansky[3] recognized that the reabsorption causes a broadening of the light emitted by resonance lamps.

In this note we report an experimental and theoretical study of the complementary effect, i.e., of resonance radiation originating in a system in which the population of the upper level of the resonance transition is made to exceed that of the lower one (inverted population system).

If the density of inverted atoms (or ions) is taken as n_2-n_1 and the spontaneous lifetime is denoted by t_{spont}, it can be shown that the effect of the inversion is to cause an amplification of radiation at frequency ν with a (exponential) gain constant $\gamma(\nu)$ given by[4]

$$\gamma(\nu)=\frac{C^2[n_2-n_1(g_2/g_1)]g(\nu)}{8\pi\nu^2 t_{\text{spont}}}=ag(\nu), \tag{1}$$

where $g(\nu)$ is the normalized line shape function of the intrinsic fluorescence, g_1 and g_2 are the level degeneracies, and a is a constant characterizing the amount of inversion and is defined by (1).

We consider next the radiation emerging from a sample of length r_0 and a distributed loss constant α. Recognizing that the light is emitted with a spectrum $g(\nu)$ we obtain, by integration, the (nonnormalized) line shape $F(\nu)$ of the emerging radiation:

$$F(\nu)=g(\nu)\frac{e^{[\gamma(\nu)-\alpha]r_0}-1}{\gamma(\nu)-\alpha}. \tag{2}$$

An immediate consequence of (2) is that for $\gamma(\nu_0)r_0\ll 1$, $F(\nu)=\text{const}\ g(\nu)$ and no change in the spectrum is to be expected. The more interesting condition is, however, when $\gamma(\nu_0)r_0\gg 1$. It is clear that if $\alpha\gg\gamma(\nu_0)$ no narrowing obtains since $F(\nu)=\text{const}\ g(\nu)$. If $\gamma(\nu_0)$ is increased to a point where it becomes comparable, yet smaller than α, $F(\nu)=\text{const}\ \{g(\nu)/[\gamma(\nu)-\alpha]\}$ which is, because of the frequency-dependent denominator, narrower than $g(\nu)$.

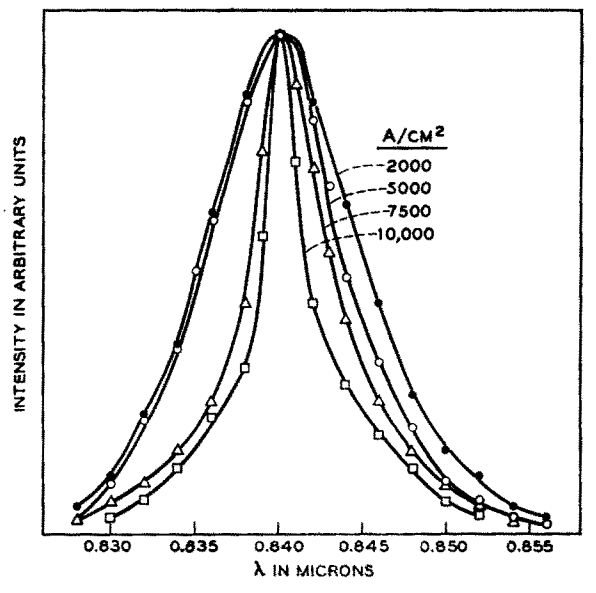

FIG. 1. Spectral profile of the recombination radiation from a GaAs p–n junction for a number of injection currents. The curves have been renormalized to a single amplitude and shifted horizontally so as to get a single peak.

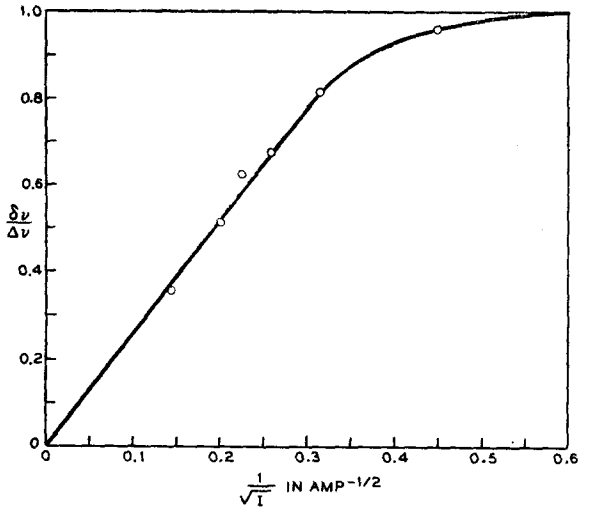

FIG. 2. Fractional narrowing as a function of injection current.

It is clear from the above that spectral narrowing, unlike oscillation, is a gradual process and that no exact narrowing condition can be given. At the same time we can summarize the observations made in the preceding section by giving the following approximate prerequisites for the "onset" of narrowing

$$\gamma(\nu_0)r_0 \approx 1, \quad \gamma(\nu_0) \sim \alpha, \tag{3}$$

where by the symbol $\sim$ we mean "of the same order of magnitude as."

The physical interpretation of this result is interesting. Conditions (3) are equivalent to stating that a photon emitted at ν_0, on the average, stimulates the emission of ~ 1 photon in a distance r_0. Photons emitted at the wings of $g(\nu)$ have, according to (1), a much smaller probability of inducing emissions so that condition (3) marks the point at which the photon distribution is beginning to vary appreciably from $g(\nu)$.

If we limit ourselves to cases where

$$\gamma(\nu_0)r_0 \gg 1, \quad \gamma(\nu_0) > \alpha, \tag{4}$$

and define $\delta\nu$ as the width at half-maximum of $F(\nu)$ and $\Delta\nu$ as the width at half-maximum of $g(\nu)$, we can solve (2) for the amount of narrowing $\delta\nu/\Delta\nu$ present at a given value of $\gamma(\nu_0)r_0$. The result is[5]

$$\delta\nu/\Delta\nu = [\gamma(\nu_0)r_0]^{-\frac{1}{2}} \text{ for a Gaussian } g(\nu) \tag{5}$$

$$\delta\nu/\Delta\nu = 0.832[\gamma(\nu_0)r_0]^{-\frac{1}{2}} \text{ for a Lorentzian } g(\nu) \tag{6}$$

from which we may conclude that the exact shape of $g(\nu)$ is not too important in determining the amount of narrowing.

The recent interest in lasers makes available a wide range of inverted population media. Unfortunately, in spite of the large number of such systems we could find only two cases where the narrowing condition $\gamma(\nu_0)r_0 > 1$ is satisfied. The first one is the large gain discharge prevailing in the Xe 3.5 μ laser[6,7] where $\gamma(\nu_0)r_0 = 12.5$ for $r_0 = 1m$. This, according to (5) should result in a $\sim$ fourfold reduction in linewidth. The practical consideration which prevented us from trying out this system was that the Doppler-broadened fluorescence has a $\Delta\nu \sim 125$ Mc/sec which makes it necessary to use optical resolution of $\sim 10^7$ to detect the narrowing.

We have chosen to check the theory on the recombination radiation emitted by forward-biased GaAs junctions. Narrowing of this radiation under conditions of high injection was first observed by Nasledov *et al.*,[8] and more recently, in a more striking fashion, by Hall *et al.*[9] and by Nathan *et al.*[10] A conservative estimate for the diode used in our experiment yielded[11]

$$\gamma(\nu_0)r_0 \sim 8$$

for

$r_0 = 0.25$ mm and an injection density of 15 000 A/cm^2 ($I = 10$ A)

$\gamma(\nu_0) = 10\alpha = 300$ cm^{-1}.[12]

In Fig. 1 are shown a number of spectral output curves obtained as the injection current was increased from 2 A to 10 A.

The narrowing is observed to set in between 2 and 4 A which, when using the estimate of $\gamma(\nu_0)r_0 = 8$ at 10 A, yields a value for $\gamma(\nu_0)r_0 \sim 2$ at the onset of narrowing, in approximate agreement with (3).

At high pumping rates, where $\gamma(\nu_0)r_0 > 1$, the fractional narrowing, according to (5) and (6), should increase as the square root of the pumping rate. In our experiment $\gamma(\nu_0)$ is proportional to the injection current I. A plot of $\delta\nu/\Delta\nu$ vs $I^{-\frac{1}{2}}$ in Fig. 2 shows a linear relationship at high currents in agreement with the theory.

It was previously proposed[13] that the narrowing mechanism described above could be used to obtain narrow noncoherent radiation sources. The square root dependence of $\delta\nu/\Delta\nu$ on the inversion shows that while the idea, in principle, is feasible we must obtain inordinately large inversions for sizeable reductions in $\delta\nu/\Delta\nu$. A survey of the laser systems available today reveals that a $\delta\nu/\Delta\nu \sim 0.1$ is probably the most we can expect at the moment.

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Criterion for Continuous Amplitude Oscillations of Optical Masers

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IN a previous report[1] we derived a set of equations describing a three-level optical maser which self-oscillates between the upper two levels. For a particular set of values for the parameters appearing in the equations a solution was found (analytically and numerically) with decay to a steady state. We wish to show here that it is possible to obtain with the same set of equations a long-time solution having a time-varying amplitude. No attempt is made in this report to find the particular analytic form of the long-time solution.

It is shown in Ref. 1 that the pertinent equations are

$$\begin{aligned} 2\omega\dot{q}_1 &= \beta_5(\sigma_2 - q_1), \\ \dot{\sigma}_2 &= \beta_2(\sigma q_1 - \sigma_2), \\ \dot{\sigma} &= -\beta_1(\sigma_0 - 1)\sigma_2 q_1 - \beta_1(\sigma - \sigma_0), \end{aligned} \tag{1}$$