

suggested in Ref. 4.

<sup>18</sup>This is basically because the lower-bound formula uses only a lower bound to the true overlap integral  $S = \langle \phi | \psi \rangle$ , so that account must be taken of the possibility  $S = 1$ , i. e., that the function  $\phi$  was actually the true

wave function  $\psi$ .

<sup>19</sup>E. A. Hylleraas, *Z. Phys.* **54**, 347 (1929).

<sup>20</sup>A number of these expectation values were previously given by J. N. Silverman, O. Platas, and F. A. Matsen, *J. Chem. Phys.* **32**, 1402 (1960).

## Coherent Enhancement of the Natural Linewidth

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When the interparticle distance of a gas of atoms is small compared with the spontaneous radiation wave length, the photon emitted by one atom may be absorbed by other atoms and may thus be trapped in the gas. The number of photons that are emitted is therefore reduced, as compared to the incoherent radiation of a gas of widely separated atoms. This photon trapping effect on the natural linewidth of the radiation is calculated and found to be large in certain cases. Some experimental aspects of observing such an effect are also discussed.

Recently Kuhn and Vaughan<sup>1</sup> reported an experimental value for the oscillator strength of the  $1^1S$ - $2^1P$  resonance transition in helium. The corresponding radiation width of the  $2^1P$  level was determined as  $1.31 \pm 0.12 \text{ m}^{-1}$  while the computed natural width<sup>2</sup> is  $0.95 \text{ m}^{-1}$ . A possible explanation of this discrepancy of about 30–40% has been suggested<sup>3</sup> in terms of coherent enhancement. The idea of coherent enhancement was first proposed and formulated by Dicke<sup>4</sup> in his discussion of the super-radiant states. More recently this effect has also been discussed<sup>5, 6</sup> for the case of two stationary atoms. These results, however, are not applicable to a many-atom gas in nonsuperradiant states. Furthermore, the latest experiment<sup>7</sup> also determined the same oscillator strength from a measurement of the  $2^1P$  lifetime to be in agreement with the computed value to  $\sim 8\%$ . Therefore it is not clear whether this particular effect of coherent enhancement is involved in these experiments.

The purpose of this paper is to investigate the conditions under which this coherence effect may be observed. We point out here that (1) according to our calculation, a large coherent enhancement of the natural linewidth does exist in certain cases, and (2) the usual linewidth experiments such as those mentioned above, in which the radiating gas is maintained in a steady state, are not suitable for observing this particular effect.

We first consider a gas which consists of  $n$  atoms in a container whose dimension is small

compared with the radiation wavelength  $\lambda$ . Generalization of the results to the case of a gas of large extent will be discussed later. The transition which gives rise to radiation of frequency  $\omega_0$  is assumed to take place between two nondegenerate  $+$  and  $-$  states of the individual atom, with corresponding eigenvalues  $\hbar\omega_0/2$  and  $-\hbar\omega_0/2$ . Following Dicke,<sup>4</sup> we assign a quantum number  $m$  as a measure of the energy of the internal states of the gas. Thus

$$m = (n_+ - n_-)/2, \quad (1)$$

where  $n_+$  and  $n_-$  are the number of atoms in  $+$  states and  $-$  states, respectively. Such a gas can be treated in analogy with a system of spin  $\frac{1}{2}$  particles. Corresponding to the total spin of the system, we now have the "cooperation number"  $r$ , whose third component is  $m$ . The assumed small size of the gas enables us to avoid the complications caused by the Doppler effect, which will be considered later. It is also assumed that collisions do not affect the internal states of the atoms, and that the interparticle distance, although small compared to  $\lambda$ , is still so large that the atoms do not interact, and the wave packet of one atom does not overlap with that of the others. Under these assumptions, the quantum number  $r$  has the important property that it remains constant throughout the radiation process.

Assuming  $r \gg 1$ , one can use the classical model

$$m = r \cos \phi(t), \quad (2)$$

in which  $\phi(t)$  changes continuously with time. The rate of radiation from the gas in the state  $\psi_{m_r}$  is then given by<sup>4</sup>

$$-\frac{d}{dt} m \hbar \omega_0 = I = I_0 [r(r+1) - m(m-1)] \\ = I_0 r^2 \sin^2 \phi(t), \quad (3)$$

where  $I_0$  is the radiation rate from a single isolated atom in the + state. We remark here that  $m$  gradually decreases with time, and this is precisely how the radiation width comes about. From Eqs. (2) and (3), it is a simple matter to show that

$$\cos \phi = \frac{(r+m_0)e^{-\alpha t} - (r-m_0)e^{\alpha t}}{(r+m_0)e^{-\alpha t} + (r-m_0)e^{\alpha t}}, \quad (4)$$

where  $m_0$  is the initial value of  $m$  at  $t=0$ , and  $\alpha \equiv I_0 r / \hbar \omega_0$ . Knowing now the time dependence of the radiation rate, we can easily determine the line shape

$$I(\omega) = 4 \frac{r+m_0}{r-m_0} \frac{1}{\alpha^2} \left( 1 + \frac{(\omega - \omega_0)^2}{\alpha^2} \right)^{-2} \quad (5)$$

and the linewidth at half-intensity points

$$\Delta\omega = 1.28\alpha = 1.28r\gamma_0, \quad (6)$$

where  $\gamma_0 = I_0 / \hbar \omega_0$  is the corresponding linewidth for the radiation from an isolated atom. In obtaining Eqs. (5) and (6), we have anticipated that, on the average, the values of  $r$  cluster around  $-m_0$ . It is observed that the line shape is different from the Lorentz shape, and that the linewidth is proportional to the cooperation number  $r$ .

Since the above calculation is made for the case of radiation from a definite (nonsuper-radiant) state  $\psi_{m_0 r}$  of the gas, an appropriate statistical ensemble average must be taken. If we assume that at  $t=0$  the excitation of the gas can be characterized by a temperature  $T$ , which is in general quite different from the temperature for the translational motion of the atoms, the canonical ensemble average value of  $m_0$  can be calculated to be

$$\overline{m_0} = (-n/2) \tanh(\hbar\omega_0/2k_B T), \quad (7)$$

where  $k_B$  is the Boltzmann constant. The average value of  $r(r+1)$ , for a fixed value of  $m$ , is given by

$$\overline{r(r+1)} = m^2 + n/2. \quad (8)$$

It can be shown<sup>8</sup> that the statistical fluctuation about these mean values becomes very small when  $n$  is sufficiently large or when  $a \equiv (\hbar\omega_0/k_B T) > 1$ , if  $n$  is not too large. Under these conditions, one can replace the average value of a function of  $r$  and  $m$  by the value of the function evaluated at

$\overline{r}$  and  $\overline{m}$ . It then follows that, in the classical model where  $r \gg 1$ , Eq. (6) together with Eqs. (7), (8) lead to a linewidth

$$\Delta\omega = 1.28\overline{r}\gamma_0 \\ = 0.64\gamma_0 [n^2 \tanh^2(\hbar\omega_0/2k_B T) + 2n]^{1/2}, \quad (9)$$

which gives the explicit dependence on  $T$  and  $n$ .

The fact that the linewidth is enhanced approximately by a factor of  $\overline{r}$  as compared with  $\gamma_0$  can be understood qualitatively in the following manner. We first note from Eqs. (3) and (8) that, for a given value of  $m_0$ , the average initial radiation rate<sup>9</sup> is given by  $\overline{I}(m_0) = (m_0 + n/2)I_0$ , which is exactly the same as the initial incoherent radiation rate  $n_+ I_0$ . We then observe from Eqs. (2) and (4) that, as  $t \rightarrow \infty$ , the average total number of emitted photons for a given  $m_0$  is  $(m_0 + \overline{r})$ . In the case of incoherent radiation, the total number of emitted photons will be  $(m_0 + n/2)$ . It is therefore natural to expect the linewidth to be enhanced by the ratio  $(m_0 + n/2)/(m_0 + \overline{r})$  as compared with the incoherent width  $\gamma_0$ . By use of the relation<sup>8</sup>  $\overline{r} \sim -m_0$  and Eq. (8), it is easy to show that the above ratio is just about  $2\overline{r}$ . Thus the enhancement of the linewidth is actually due to the fact that the states  $\psi_{m_0 r}$  have less photons to emit than in the incoherent case. In fact,  $(n/2 - r)$  can be regarded as the number of photons trapped in the state  $\psi_{m_0 r}$ . This photon trapping arises from the emission and reabsorption of the photon by the many atoms spaced closely together.

It often happens that the excitation of the gas cannot be described by a Boltzmann distribution of atoms in the excited levels. The temperature  $T$  in Eqs. (7), (9) then loses its meaning. However, as long as we assume that the statistical fluctuations about the mean values are small, we can still write, based on Eq. (6),

$$I(\omega) \propto \frac{\overline{r} + \overline{m_0}}{\overline{r} - \overline{m_0}} \left( \frac{1}{\overline{\alpha}} \right)^2 \left( 1 + \frac{(\omega - \omega_0)^2}{(\overline{\alpha})^2} \right)^{-2}, \quad (10)$$

$$\text{and } \Delta\omega = 1.28 \overline{\alpha} = 1.28 \gamma_0 \overline{r}. \quad (11)$$

Since Eq. (8) is still valid, we see that both the intensity and the linewidth can be expressed in terms of the single parameter  $\overline{m_0}$ , which is a measure of the degree of excitation of the gas. By combining Eqs. (10) and (11), the relative radiation intensity can be expressed as a function of the linewidth. If one changes the experimental parameter, such as the power of the excitation source or the pressure of the gas,  $\overline{m_0}$  will change. An experimental verification of the dependence of the relative intensity on the linewidth as  $\overline{m_0}$  is varied may be taken as confirmation of the coherent enhancement. This is in contrast to the case of incoherent radiation in which the radiation intensity varies with the experimental parameters

such as  $\bar{m}_0$  and  $n$  while the natural linewidth remains unchanged.

So far we have been assuming that the wavelength of the radiation  $\lambda$  is large compared with the dimension of the system. This is justified, for example, when the radiating system is a set of proton spins in a magnetic field. It remains to be determined whether the above results and conclusions can be applied to a system much larger than the wavelength. To get some insight into this question, we examine also the case of two stationary atoms,<sup>5,6</sup> one of which is in the + state and the other in the - state. Since the direct dipole-dipole interaction between the two atoms does not affect the linewidth, we neglect it for simplicity. The radiation intensity distribution is then found to be

$$I(\omega_k) \sim \frac{\gamma}{(\omega_0 - \omega_k)^2 + \gamma^2} + \frac{\gamma'}{(\omega_0 - \omega_k)^2 + \gamma'^2}, \quad (12)$$

where

$$\gamma = \gamma_0 \left( 1 + \frac{\sin kL}{kL} \right), \quad \gamma' = \gamma_0 \left( 1 - \frac{\sin kL}{kL} \right),$$

and  $L$  is the interparticle distance. It is seen from Eq. (12) that the coherence effect, represented by the term  $(\sin kL)/kL$ , is important only when  $kL < \pi$ . As  $kL \rightarrow 0$ , the first term in Eq. (12) represents the contribution of the "coherent photons" while the second term represents the trapped photons. When  $kL > \pi$ , the coherence becomes negligible and the two atoms act incoherently. It is therefore reasonable to expect that for a large system our results will still hold true qualitatively if we interpret  $n$  as the number of particles within a volume of  $\sim (\lambda/2)^3$ . This implies that the large system may be considered as an incoherent collection of small systems, each of which consists of  $n$  particles participating coherently in the radiation process.

Another question is the relaxation effect of random thermal motion and collisions of the particles on the coherence. To discuss this, let us again consider the case of two randomly moving particles confined to a small box of linear dimension  $L$ , with one of the particles in the excited + state. It can be shown explicitly<sup>10</sup> that, for small values of  $kL$ , the radiation intensity distribution is still very similar to that of Eq. (12). The only modi-

fication is that a small part of the "coherent photons" has been transferred from the high peak of width  $\sim 2\gamma_0$  corresponding to the first term of Eq. (12) to a broad and shallow background distribution of width  $2\pi\bar{v}/L$  due to the random motion of average speed  $\bar{v}$ . The ratio of the number of photons in the shallow background to that under the high peak is  $0.02k^2L^2$ , which is small and generally independent of  $\bar{v}$ . Thus we conclude that random motion does not affect the coherent radiation from the two particles in a small box. This conclusion will not be changed, of course, if one of the particles has gone out of the box, provided there is another particle like that entering from outside to take its place. Physically this means that as long as there are, on the average,  $n$  particles closely spaced together within half a wavelength or so, photon trapping and hence an enhancement of the radiation linewidth will result, irrespective of the random motion of the particles.

Finally we again emphasize that it is the change of state of the whole gas as it radiates that gives rise to the natural linewidth. In the usual experiments, however, the radiating gas is maintained in a steady state. If the particles were far apart and radiated independently of each other, this steady-state nature would not affect the natural linewidth. This is because the process of radiating energy away by some particles is completely unrelated to the process of energy replenishing by an external power source through some other independently acting particles. On the other hand, when the particles are close together and radiate as a coherent whole, the steady-state experiments are clearly not suitable for measuring the natural linewidth. In order to observe such enhancement, it may be necessary to perform an experiment in which the gas radiates and de-excites from an initial state. One may, for example, pass a beam of excited atoms through a hole on the wall of the gas container into high vacuum and measure the radiation there.

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<sup>4</sup>R. H. Dicke, Phys. Rev. 93, 99 (1954).

<sup>5</sup>M. J. Stephen, J. Chem. Phys. 40, 669 (1964).

<sup>6</sup>D. A. Hutchinson and H. F. Hameka, J. Chem. Phys. 41, 2006 (1964).

<sup>7</sup>W. L. Williams and E. S. Fry, Phys. Rev. Letters 20, 1335 (1968).

<sup>8</sup>Using the notation  $\langle \dots \rangle$  instead of the bar for average, we have  $\langle m \rangle = -\frac{1}{2}n \tanh \frac{1}{2}a$ , and  $\langle m^2 \rangle = \frac{1}{4}n(n-1) \tanh^2 \frac{1}{2}a + \frac{1}{4}n$ , so that

$$\langle m^2 \rangle - \langle m \rangle^2 / \langle m \rangle^2 = (1 - \tanh^2 \frac{1}{2}a) / n \tanh^2 \frac{1}{2}a$$

and

$$\begin{aligned} \langle r(r+1) \rangle^2 - \langle [r(r+1)]^2 \rangle / \langle r(r+1) \rangle^2 \\ = \frac{4(n-1)(1 - \tanh^2 \frac{1}{2}a)}{n[(n-1) \tanh^2 \frac{1}{2}a + 3]^2}. \end{aligned}$$

We can also show that

$$\frac{\langle r(r+1) \rangle - \langle m^2 \rangle}{\langle r(r+1) \rangle} = \frac{2}{(n-1) \tanh^2 \frac{1}{2}a + 3}$$

and therefore  $\langle r \rangle + \langle m \rangle \approx 0$  for large  $n$  or small  $T$ .

<sup>9</sup>That this initial average rate is the same as the initial incoherent rate is because some  $\psi_{m_0 r}$  states are constructively coherent while other  $\psi_{m_0 r}$  states are destructively coherent. However, all the particles do participate coherently in the radiation from each of these  $\psi_{m_0 r}$  states so that  $r$  remains constant throughout the radiation process. This is in contrast to the incoherent case, in which  $r(r+1)$  changes like  $[m^2 + (n/2)]$  during the radiation process.

<sup>10</sup>Y. C. Lee and D. L. Lin, Phys. Rev. **183**, (1969); following paper. In this reference, the different time scales for the observation of the "coherent photons" and the "trapped photons" are also discussed.

## Coherent Radiation from Atoms in Random Motion

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The effect of the random motion of atoms in a gas on the coherence of the radiation from the gas is investigated. It is found that, as long as the spontaneous radiation wavelength is sufficiently greater than the interparticle distance so that a coherence effect exists for stationary atoms, the actual random motion of the atoms can only cause a minor modification.

### I. INTRODUCTION

In a recent paper, Lee and Lin<sup>1</sup> have shown that the natural linewidth of the spectral line from a many-atom gas is in general enhanced due to the coherence effect, and that large enhancement can be observed under certain circumstances. These conclusions are reached by a generalization of the results that hold true for a two-atom system. Stephen<sup>2</sup> and Hutchinson and Hamka<sup>3</sup> have independently investigated the effect on the lifetime of an excited atom due to the presence of another identical particle in its ground state. The explicit calculations made in Refs. 2 and 3 show clearly that the coherence effect on the linewidth does not depend on the direct interaction between the atoms. For this reason, we have re-examined the two-atom problem, ignoring the direct interaction, and obtained a simpler form for the line shape.<sup>1</sup>

$$I(\omega_k) \sim \left\{ \frac{\gamma}{(\omega_0 - \omega_k)^2 + \gamma^2} + \frac{\gamma'}{(\omega_0 - \omega_k)^2 + (\gamma')^2} \right\}, \quad (1)$$

$$\text{where } \gamma = \gamma_0 \left( 1 + \frac{\sin kL}{kL} \right), \quad \gamma' = \gamma_0 \left( 1 - \frac{\sin kL}{kL} \right),$$

with  $\gamma_0$  representing the linewidth for an isolated atom,  $L$  the interparticle distance, and  $k$  the wave number of the radiation. It is observed that when  $kL < \pi$ , the first term in (1) exhibits the coherence broadening, yielding a width of  $2\gamma_0$  in the limit of  $kL \rightarrow 0$ , while for  $kL > \pi$ , both terms are alike, and the radiation becomes incoherent for large  $kL$ .

The above result is obtained for two stationary noninteracting particles. It is then natural to ask whether or not the random motion of the particles will destroy the coherence. We shall, in this paper, investigate this question and show that the random motion of the radiating atoms has very little effect on the coherently enhanced width as long as  $kL < \pi$ .

Our calculation is based on the general method of Heitler and Ma,<sup>4</sup> which is extended here to include the random motion of the atoms. We first formulate the problem in Sec. II and then develop the method of calculation in Sec. III. The results