

The Heitler and Ma approach to resonance fluorescence of N two-level atoms in a weak electromagnetic field

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1979 J. Phys. A: Math. Gen. 12 1739

(<http://iopscience.iop.org/0305-4470/12/10/020>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.252.86.83

The article was downloaded on 30/05/2010 at 19:04

Please note that [terms and conditions apply](#).

The Heitler and Ma approach to resonance fluorescence of N two-level atoms in a weak electromagnetic field

R Pyżalski

Institute of Chemistry, A Mickiewicz University, 60-780 Poznań, Grunwaldzka 6, Poland

Received 31 August 1978

Abstract. The spectral intensity distribution of resonance fluorescence radiation emitted by N two-level atoms is calculated for the case of a weak electromagnetic field. Using these results the spectrum is discussed in terms of the number of atoms, their localisation and the orientation of the transition dipole moments. It is shown that the spectrum can be expressed as a sum of Lorentzian lines only in some specific cases. The general condition describing these cases is given.

1. Introduction

The theory of resonance fluorescence of one atom placed in a weak electromagnetic field (Weisskopf 1931, Heitler 1954) can be applied to an N atom system only in the case in which each atom interacts with the radiation independently of the others. This assumption is equivalent to a neglect of photon exchange between the atoms. It is well known, however, that photon exchange is essential in the processes of interaction between many atoms and an electromagnetic field. As a consequence, the N atom system should be considered as one quantum system (Dicke 1954).

Calculations in the case of resonance fluorescence of two two-level atoms in a weak electromagnetic field show that the photon exchange causes dipole-dipole interactions between the atoms and, hence, characteristic changes in the emission spectrum. Two Lorentzian lines originating from the emission of symmetric and anti-symmetric two-atoms states are split proportionally to the interaction energy of the atoms (Chang and Stehle 1971, Fontana and Hearn 1967, Hearn and Fontana 1969). Thus the resonance fluorescence spectrum is similar to the spontaneous emission spectrum (Stephen 1964, Czarnik and Fontana 1969, Lehmborg 1970) but because the initial conditions are different the lines have different heights (Czarnik and Fontana 1969).

In the present paper the intensity distribution of radiation emitted in resonance fluorescence is calculated for the case of N two-level, identical atoms with definite localisations. It is assumed that the atoms interact with a weak electromagnetic field and that only one atom in the N atom system can be excited. The intensity distribution is obtained by using the Heitler and Ma method of solving the time-dependent Schrödinger equation (Heitler and Ma 1949, Heitler 1954). However, the rotating wave approximation (RWA) which is usually applied along with the Heitler and Ma method (cf Agarwal 1974) is not used. The anti-resonance components of the interaction Hamiltonian, neglected in the RWA, are necessary to obtain the correct form of the cooperative energy shifts (Knight and Allen 1973, Milonni and Knight 1974) from which the spectral line shifts are determined.

2. Solution of Schrödinger equation†

The \hat{H}_s Hamiltonian of the N two-level atoms interacting with an electromagnetic field has the form

$$\hat{H}_s = \hat{H}_0 + \hat{H} \quad (1)$$

where $\hat{H}_0 = \hat{H}_A + \hat{H}_F$ and

$$\hat{H}_A = \sum_{j=1}^N \frac{1}{2} k_0 \hat{\sigma}_j^z \quad (2)$$

is the Hamiltonian of N isolated atoms with excitation energy k_0 . $\hat{\sigma}_j^z$ is the operator of the z -th component of energetic spin of j -th atom.

Next

$$\hat{H}_F = \sum_{\mathbf{k}} k \hat{a}_{\mathbf{k}}^{\dagger} \hat{a}_{\mathbf{k}} \quad (3)$$

is the Hamiltonian of the free field, where $k = |\mathbf{k}|$ and \mathbf{k} is the wave vector of the electromagnetic field.

The operator \hat{H} is the Hamiltonian of the interaction between the atoms and the field. In the dipole approximation

$$\hat{H} = \sum_{j=1}^N \hat{H}_j \quad (4)$$

$$\hat{H}_j = \sum_{\mathbf{k}} (g_{kj} \hat{a}_{\mathbf{k}} + g_{kj}^* \hat{a}_{\mathbf{k}}^{\dagger}) (\hat{\sigma}_j^{-} + \hat{\sigma}_j^{+})$$

where the coupling constant g_{kj} has the form

$$g_{kj} = i(2\pi k/V)^{1/2} d e^{i\mathbf{k}\cdot\mathbf{r}_j} \mathbf{d}_j \cdot \mathbf{e}_{\mathbf{k}} \quad (5)$$

and $d\mathbf{d}_j$ is a dipole transition moment for two levels of the j -th atom. \mathbf{d}_j denotes the versor of the dipole moment and $|\mathbf{d}_j| = 1$. d is the modulus of the dipole moment which is the same for all atoms. $\mathbf{e}_{\mathbf{k}}$ is a polarisation versor of the electromagnetic wave. \mathbf{r}_j is a position vector of the electron in the j -th atom.

The sum over the wave vector \mathbf{k} in (4) includes the sum over the photon polarisation. Commutation rules of spin and field operators are as follows

$$\begin{aligned} [\hat{\sigma}_j^{-}, \hat{\sigma}_j^{+}] &= -\hat{\sigma}_j^z \delta_{jj'} \\ [\hat{\sigma}_j^{\pm}, \hat{\sigma}_j^z] &= \mp 2\hat{\sigma}_j^{\pm} \delta_{jj'} \\ [\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}'}^{\dagger}] &= \delta_{\mathbf{k}\mathbf{k}'} \end{aligned} \quad (6)$$

The other commutators are equal to zero.

The time-dependent Schrödinger equation for the system of interest has the form

$$i \frac{\partial |\chi(t)\rangle}{\partial t} = \hat{H}_s |\chi(t)\rangle. \quad (7)$$

The Heitler and Ma (1949) method will be used to solve equation (7). For this purpose

† $\hbar = c = m = 1$ units are used.

the time-dependent state vector $|\chi(t)\rangle$ is given by the expression

$$|\chi(t)\rangle = \sum_n b_n(t) e^{-iE_n t} |\chi_n\rangle \tag{8}$$

where $|\chi_n\rangle$ are the eigenstates of the Hamiltonian \hat{H}_0 with energies E_n . We consider only the following states:

(i) The initial state with energy E_0 . All atoms are in ground states and there are n_k photons in the field.

$$|0\rangle = \prod_{i=1}^N |i^0\rangle |n_k, 0_{k'}\rangle. \tag{9}$$

(ii) The real intermediate states with energies E_{ik} . Only one atom is excited and one photon is absorbed from the field

$$|ik\rangle = |i^e\rangle \prod_{j \neq i}^N |j^0\rangle |n_k - 1, 0_{k'}\rangle. \tag{10}$$

(iii) The virtual intermediate states for which the energy is not conserved (i.e. with energy $E_{ijkk'}$ differing from the energy of other states by much more than the level's width). Two atoms are excited and there is an additional photon with energy k' in the field

$$|ijkk'\rangle = |i^e\rangle |j^e\rangle \prod_{l \neq i, j}^N |l^0\rangle |n_k - 1, 1_{k'}\rangle. \tag{11}$$

(iv) The final state with energy $E_{fkk'}$. All atoms are in the ground state again and there is an additional photon with energy k' in the field

$$|fkk'\rangle = \prod_{i=1}^N |i^0\rangle |n_k - 1, 1_{k'}\rangle. \tag{12}$$

In all these cases $|i^0\rangle$ and $|i^e\rangle$ are the vectors of the ground and excited states of the i -th atom, respectively. $|n_k, n_{k'}\rangle$ is the state vector of the field in the Fock representation in which there are $n_k + n_{k'}$ photons with energies k and k' and polarisations e_k and $e_{k'}$, respectively.

The states described by the vectors in (9), (10) and (12) have been applied many times in problems of interaction between atoms and a weak field (Fontana and Hearn 1967, Hearn and Fontana 1969, Czarnik and Fontana 1969, Hutchinson and Hamerka 1964, Freedhoff and van Kranendonk 1967). The virtual states given in (11) are considered in order to include in the calculations the anti-resonant components $\hat{a}_k^+ \hat{\sigma}_j^+$ and $\hat{a}_k \hat{\sigma}_j^-$ of the interaction Hamiltonian. These components are often neglected in the case of a weak field because it is assumed they are insignificant (Louisell 1964). The anti-resonant components cannot be neglected when cooperative energy shifts occur in the calculations (Knight and Allen 1973, Agarwal 1973, 1974) and the rotating wave approximation has not been used in those cases (cf, for example, Milonni and Knight 1974). Thus calculations in the present paper differ from the preceding ones based on the Heitler and Ma method and they do not require an additional assumption about atom interaction (cf Fontana and Hearn 1967 and the discussion of their paper by Chang and Stehle 1971).

The definition of states $|\chi_n\rangle$ in (8) allows one to write an explicit form of the equations for the probability amplitude of the states $|\chi_n\rangle$. According to the Heitler and

Ma method (1954) we have

$$b_n(t) = \frac{i}{2\pi} \int_{-\infty}^{+\infty} dE \left[E - E_0 + \frac{i}{2} \Gamma(E) \right]^{-1} W_n(E) e^{i(E_n - E)t} \tag{13}$$

and $W_0 \equiv 1$. The quantities $\Gamma(E)$ and $W_n(E)$ are given by equations

$$\sum_n H_{mn} W_n(E) = \begin{cases} -\frac{i}{2} \Gamma(E) & m = 0 \\ (E - E_m) W_m(E) & m \neq 0 \end{cases} \tag{14}$$

where $H_{mn} = \langle \chi_m | \hat{H} | \chi_n \rangle$.

These equations contain the initial condition, i.e. for $t = 0$ only the state $|0\rangle$ is occupied.

Making use of the properties of the state vectors (9–12) we rewrite (14) in matrix form

$$(E - E_{fkk'}) W_{fkk'} = \mathbf{H}_{fkk'}^+ \mathbf{W} \tag{15}$$

$$\mathbf{A} \mathbf{W} = \mathbf{H}_0 \tag{16}$$

$$-\frac{i}{2} \Gamma(E) = \sum_k \mathbf{H}_0^+ \mathbf{W} \tag{17}$$

where \mathbf{W} , \mathbf{H}_0 and $\mathbf{H}_{fkk'}$ are column matrices

$$\mathbf{W} = \begin{pmatrix} W_{1k} \\ W_{2k} \\ \vdots \\ W_{Nk} \end{pmatrix} \quad \mathbf{H}_0 = \begin{pmatrix} H_{1k,0} \\ H_{2k,0} \\ \vdots \\ H_{Nk,0} \end{pmatrix} \quad \mathbf{H}_{fkk'} = \begin{pmatrix} H_{1k, fkk'} \\ H_{2k, fkk'} \\ \vdots \\ H_{Nk, fkk'} \end{pmatrix} \tag{18}$$

and \mathbf{A} is a square matrix ($N \times N$) of the following elements

$$(\mathbf{A})_{ij} = \left(E - E_{ik} + \frac{i}{2} \sum_{l \neq i} \gamma_{ik,il}^{(l)} \right) \delta_{ij} + \frac{i}{2} (\gamma'_{ik,jk} + \gamma_{ik,jk}). \tag{19}$$

The quantities γ_{mn} are

$$-\frac{i}{2} \gamma_{ik,ik}^{(l)} = \sum_{k'} H_{ik,ilk'k'} H_{ilk',ik} \zeta(E - E_{ilk'}) \tag{20}$$

$$-\frac{i}{2} \gamma'_{ik,jk} = \sum_{k'} H_{ik,ijk'k'} H_{ijk',jk} \zeta(E - E_{ijk'}) \tag{21}$$

$$-\frac{i}{2} \gamma_{ik,jk} = \sum_{k'} H_{ik, fkk'k'} H_{fkk',jk} \zeta(E - E_{fkk'}) \tag{22}$$

where $\zeta(x) = P(1/x) - i\pi\delta(x)$ and P denotes the principal value in the Cauchy sense.

The equations (15–17) are equivalent to the Schrödinger equation (7) for resonance fluorescence. Their solutions can be also written in matrix form

$$\mathbf{W} = \mathbf{A}^{-1} \mathbf{H}_0 \tag{23}$$

$$(E - E_{fkk'}) W_{fkk'} = \mathbf{H}_{fkk'}^+ \mathbf{A}^{-1} \mathbf{H}_0 \tag{24}$$

$$-\frac{i}{2}\Gamma(E) = \sum_k \mathbf{H}_0^+ \mathbf{A}^{-1} \mathbf{H}_0 \tag{25}$$

if only one assumes that the matrix \mathbf{A} is non-singular.

3. Intensity distribution of emitted radiation

Using the solutions (23–25) and integral transformation (13) it would be possible to obtain the time-dependent state vector (8) but the vector state is not necessary to calculate the emission spectrum because this spectrum is given by the probability of the occupation of the final state for time $t \rightarrow \infty$. According to the Heitler and Ma method (1954) we write

$$|b_{fkk'}(\infty)|^2 = \left| \frac{(E - E_{fkk'})W_{fkk'}}{E - E_0 + (i/2)\Gamma(E)} \right|_f^2 \tag{26}$$

The solitary index ‘ f ’ in this and the next equations denotes the quantity that should be calculated for $E = E_{fkk'}$.

Now we define two Hermite matrices

$$\begin{aligned} \mathbf{F}_{fkk'} &= \mathbf{H}_{fkk'} \mathbf{H}_{fkk'}^+ \\ \mathbf{F}_0 &= \mathbf{H}_0 \mathbf{H}_0^+ \end{aligned} \tag{27}$$

and use solution (24) to write the occupation probability (26) in another form

$$|b_{fkk'}(\infty)|^2 = \frac{\text{Tr}[(\mathbf{F}_{fkk'} \mathbf{A}_f^{-1})^+ (\mathbf{A}_f^{-1} \mathbf{F}_0)]}{|E_{fkk'} - E_0 + (i/2)\Gamma(E_{fkk'})|^2} \tag{28}$$

where Tr means the trace of a matrix.

From (28) we derive the intensity distribution of the emitted radiation by integration over the orientation and summation over the polarisation of emitted and incoming photons and by summation over the frequencies of the incoming photons. Here we assume a continuous spectrum of incoming radiation. Thus the intensity distribution has the form

$$I(\Delta) = \gamma^2 \text{Tr}[(\mathbf{F} \mathbf{A}_f^{-1})^+ (\mathbf{A}_f^{-1} \mathbf{F})] \tag{29}$$

where

$$\mathbf{A}_f = \gamma \left(\Delta \mathbf{1} - \mathbf{V} + \frac{i}{2} \mathbf{F} \right) \tag{30}$$

$$(\mathbf{F})_{ij} = \{(\mathbf{d}_i \cdot \mathbf{d}_j)j_0(k_0 \mathbf{R}_{ij}) + \frac{1}{2}[3\mathbf{R}_{ij}^{-2}(\mathbf{d}_i \cdot \mathbf{R}_{ij})(\mathbf{d}_j \cdot \mathbf{R}_{ij}) - \mathbf{d}_i \cdot \mathbf{d}_j]j_2(k_0 \mathbf{R}_{ij})\}(1 - \delta_{ij}) + \delta_{ij} \tag{31}$$

$$(\mathbf{V})_{ij} = -\frac{1}{2}\{(\mathbf{d}_i \cdot \mathbf{d}_j)j_{-1}(k_0 \mathbf{R}_{ij}) + \frac{1}{2}[3\mathbf{R}_{ij}^{-2}(\mathbf{d}_i \cdot \mathbf{R}_{ij})(\mathbf{d}_j \cdot \mathbf{R}_{ij}) - \mathbf{d}_i \cdot \mathbf{d}_j]j_{-3}(k_0 \mathbf{R}_{ij})\}(1 - \delta_{ij}). \tag{32}$$

The quantity γ is the decay rate of one isolated atom

$$\gamma = \frac{4}{3}|d|^2 k_0^3. \tag{33}$$

$\Delta = (k_0 - k')/\gamma$ is a variable determining the deviation from resonance; $\mathbf{1}$ is the unit matrix ($N \times N$); $j_n(x)$ are spherical Bessel functions of n -th order; $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_j$, where $\mathbf{R}_i, \mathbf{R}_j$ are the position vectors of the i -th and j -th atoms and $\mathbf{R}_{ij} = |\mathbf{R}_{ij}|$

In the result (29) the non-cooperative energy shifts have been neglected. They arise in resonance fluorescence just as in the spontaneous emission of N two-level atoms (cf Saunders and Bullough 1973). These shifts do not modify the spectrum because the energies of all intermediate states are shifted by the same value. Moreover, we have assumed that the size of atoms is much smaller than the resonance wavelength of the radiation and hence

$$\exp(ik \cdot r_i) \approx \exp(ik \cdot R_i). \quad (34)$$

This approximation is called the relative dipole approximation.

4. Properties of the emission spectrum

The expression (29) permits an easy discussion of the emission line shape in resonance fluorescence of N atoms. We rewrite (29) in a convenient form

$$I(\Delta) = \text{Tr}\{[\mathbf{F}^{-1}(\Delta\mathbf{1} - \mathbf{V})]^2 + \frac{1}{4}\mathbf{1}\}^{-1} \quad (35)$$

which is equivalent to (29) if matrix \mathbf{F} is non-singular.

The intensity distribution (35) is an invariant of the unitary transformation carried jointly over the matrices \mathbf{F} and \mathbf{V} , i.e. instead of (35) we may write

$$I(\Delta) = \text{Tr}\{[\tilde{\mathbf{F}}^{-1}(\Delta\mathbf{1} - \tilde{\mathbf{V}})]^2 + \frac{1}{4}\mathbf{1}\}^{-1} \quad (36)$$

where

$$\tilde{\mathbf{X}} = \mathbf{M}\mathbf{X}\mathbf{M}^+ \quad (37)$$

and \mathbf{M} is any unitary matrix ($\mathbf{M}^+ = \mathbf{M}^{-1}$).

The matrices \mathbf{F} and \mathbf{V} are normal (i.e. real and symmetric). If we set up a suitable matrix \mathbf{M} it is possible to diagonalise matrix \mathbf{F} or \mathbf{V} . In the particular case matrices \mathbf{F} and \mathbf{V} commute, i.e. when

$$[\mathbf{F}, \mathbf{V}] = \mathbf{0} \quad (38)$$

the matrix \mathbf{M} may diagonalise both \mathbf{F} and \mathbf{V} .

It can be proved that the unitary transformation (37) is equivalent to a change of the real intermediate states (from states $|ik\rangle$ to others which are their linear combination). These new intermediate states are simply connected with matrix \mathbf{M} :

$$|\chi'_k\rangle = \mathbf{M}^*|\chi_k\rangle \quad (39)$$

where

$$|\chi_k\rangle^T = (|1k\rangle|2k\rangle \dots |Nk\rangle). \quad (40)$$

Thus if matrices \mathbf{F} and \mathbf{V} commute and suitable real intermediate states are set up then the intensity distribution may be expressed by two diagonal matrices $\tilde{\mathbf{F}}_D$ and $\tilde{\mathbf{V}}_D$ (the index 'D' denotes the diagonal form of the matrix). Therefore

$$I(\Delta) = \text{Tr}\{[\tilde{\mathbf{F}}_D^{-1}(\Delta\mathbf{1} - \tilde{\mathbf{V}}_D)]^2 + \frac{1}{4}\mathbf{1}\}^{-1} \quad (41)$$

or

$$I(\Delta) = \sum_{i=1}^N \frac{(\tilde{\mathbf{F}}_D)_{ii}^2}{[\Delta - (\tilde{\mathbf{V}}_D)_{ii}]^2 + \frac{1}{4}(\tilde{\mathbf{F}}_D)_{ii}^2}. \quad (42)$$

Each component of the sum (42) is a Lorentzian function of the variable Δ and describes the intensity distribution of radiation emitted by one intermediate state defined by (39).

Let us consider a case when matrices \mathbf{F} and \mathbf{V} do not commute. Then only one of them may be diagonalised by the matrix \mathbf{M} . The equation (42) is invalid and it is impossible to express the intensity distribution $I(\Delta)$ by the sum of the Lorentzian components. Thereby we have an important result: commutator (38) is a condition which determines whether the spectrum is a sum of Lorentzian lines or not.

Now we give more detailed properties of the spectrum in both cases. If $[\mathbf{F}, \mathbf{V}] = \mathbf{0}$, the spectrum consists of N Lorentzian lines. The i -th of them is shifted in relation to the line of the isolated atom by the value of $(\tilde{\mathbf{V}}_D)_{ii}$. Its halfwidth is equal to $(\tilde{\mathbf{F}}_D)_{ii}$. The sum of all line shifts is equal to zero.

$$\sum_{i=1}^N (\tilde{\mathbf{V}}_D)_{ii} = \text{Tr } \mathbf{V} = 0 \tag{43}$$

and the sum of the halfwidths is N times greater than the half width of one isolated atom

$$\sum_{i=1}^N (\tilde{\mathbf{F}}_D)_{ii} = \text{Tr } \mathbf{F} = N. \tag{44}$$

Both $(\tilde{\mathbf{V}}_D)_{ii}$ and $(\tilde{\mathbf{F}}_D)_{ii}$ have a cooperative character. $(\tilde{\mathbf{V}}_D)_{ii}$ is the cooperative energy shift (and in the case of interest it is also a cooperative line shift). $(\tilde{\mathbf{F}}_D)_{ii}$ is the cooperative line width.

If $[\mathbf{F}, \mathbf{V}] \neq \mathbf{0}$, the intensity distribution is expressed by N^2 positive defined rational functions which are non-Lorentzian ones. The detailed form of these functions may be obtained by writing out expression (36) in which the matrix \mathbf{F} is diagonal.

Now we will consider some special cases. First, we assume that all interatomic distances are much greater than the resonance wavelength:

$$k_0 R_{ij} \gg 1. \tag{45}$$

Then the matrices \mathbf{F} and \mathbf{V} commute and have simple forms

$$\mathbf{F} = \mathbf{1} \quad \mathbf{V} = \mathbf{0}. \tag{46}$$

The intensity distribution (42) is

$$I(\Delta) = N/(\Delta^2 + \frac{1}{4}) \tag{47}$$

i.e. it is expressed by N identical Lorentzian lines which are characteristic for the resonance fluorescence of one isolated atom (Weisskopf 1931, Heitler 1954).

In the opposite case

$$k_0 R_{ij} \ll 1 \tag{48}$$

and the system fulfilling this condition is called a 'small sample'. Now we have

$$(\mathbf{F})_{ij} \approx 1 \tag{49}$$

$$(\mathbf{V})_{ij} \approx (k_0 R_{ij})^{-3} [1 - 3R_{ij}^{-2} (\mathbf{d} \cdot \mathbf{R}_{ij})^2] (1 - \delta_{ij}) \tag{50}$$

and in (50) the orientations of the transition dipole moments of all atoms are assumed to be parallel to the versor \mathbf{d} . Equation $[\mathbf{F}, \mathbf{V}] = \mathbf{0}$ is satisfied only in specific cases. This is easily seen from its equivalent form (together with (49))

$$\sum_j (\mathbf{V})_{jl} = \sum_j (\mathbf{V})_{ji} \quad i \neq l. \tag{51}$$

To obtain the intensity distribution we cannot apply (35) because the matrix \mathbf{F} is singular. The equation (29) however is still correct since the matrix \mathbf{A} is always non-singular. If we set up a suitable real intermediate state so that the matrix \mathbf{F} is diagonal then the intensity distribution has the form

$$I(\Delta) = N^2 / (|\Delta \mathbf{1} - \hat{\mathbf{V}}| / |(\Delta \mathbf{1} - \hat{\mathbf{V}})_{(11)}|)^2 + N^2 / 4 \quad (52)$$

where $|\mathbf{X}|$ and $|(\mathbf{X})_{(11)}|$ denote the determinant and complementary minor of element (11) of matrix \mathbf{X} , respectively. This result gives the spectrum of the radiation emitted from the totally symmetric state of N atoms, i.e. from the state

$$(1/\sqrt{N}) \sum_{i=1}^N |ik\rangle. \quad (53)$$

Other intermediate states do not radiate. This property is characteristic for small samples in spontaneous emission (Dicke 1954) as well as in resonance fluorescence processes.

The shape of the emission line for the small sample, given by (52), depends on the number of atoms. For one and two atoms we obtain Lorentzian lines

$$I(\Delta)_{N=1} = 1 / (\Delta^2 + \frac{1}{4}) \quad (54)$$

$$I(\Delta)_{N=2} = 4 / [(\Delta - V)^2 + 1] \quad (55)$$

where $V = (\mathbf{V})_{12}$.

The spectrum (55) is similar to Stephen's (1964) result for the spontaneous emission of two atoms.

In the three-atom system the line may be of non-Lorentzian shape. From (52) we have

$$I(\Delta)_{N=3} = 9 \left\{ \left[3 \frac{\Delta^3 - \Delta(V_{12}^2 + V_{13}^2 + V_{23}^2) - 2V_{12}V_{13}V_{23}}{(\Delta + V_{12} + V_{13} + V_{23})^2 + 2(\Delta^2 - V_{12}^2 - V_{13}^2 - V_{23}^2)} \right]^2 + \frac{9}{4} \right\}^{-1} \quad (56)$$

where $V_{ij} = (\mathbf{V})_{ij}$.

Condition (51) is fulfilled only in the particular cases when $V_{12} = V_{13} = V_{23} \equiv V$ and Lorentzian line is obtained

$$I(\Delta)_{N=3} = 9 / [(\Delta - 2V)^2 + 9/4]. \quad (57)$$

It is worth noting that the equality $V_{12} = V_{13} = V_{23}$ may be an effect of the specific arrangement of the atoms in space. Thus a change in the mutual localisation of the atoms may entail an essential modification of the line shape. It was shown by Stephen (1964) that the quantity $\gamma(\mathbf{V})_{ij}$ is the energy of the dispersion interaction between the atoms in the excited state and atoms in the ground state. According to (55) the dispersion interaction in a two-atom system causes a shift of the line only. A greater number of atoms may also change the Lorentzian line shape and give lines which are described by other rational functions. To visualise the difference between the two cases it is sufficient to compare equations (55) and (56) and to note that the intensity distribution (56) may have several maxima while the distribution (55) has only one maximum.

It is necessary to point out that results for a small sample lose their meaning when the parameter $k_0 R_{ij}$ tends to zero since the energy of the dispersion interaction tends to infinity owing to the breaking of the dipole approximation (Knight and Allen 1973).

As a final case we may consider a system with interatomic distances comparable with the resonance wavelength

$$k_0 R_{ij} \approx 1. \tag{58}$$

As in the case of a small sample, only the two-atom system has the Lorentzian spectrum (here we neglect the trivial case $N = 1$). The intensity distribution has the form

$$I(\Delta)_{N=2} = \frac{(1+F)^2}{(\Delta - V)^2 + \frac{1}{4}(1+F)^2} + \frac{(1-F)^2}{(\Delta + V)^2 + \frac{1}{4}(1-F)^2} \tag{59}$$

where $V = (V)_{12}$ and $F = (F)_{12}$.

Two Lorentzian lines are connected with the emission from the symmetric and anti-symmetric states of two atoms, respectively. The half width of these lines are $1 + F$ and $1 - F$ respectively. Expression (59) is identical to that of Chang and Stehle (1971).

An analogous result for the spontaneous emission of two two-level atoms (Lehmberg 1970) is

$$I(\Delta) = \frac{1+F}{(\Delta - V)^2 + \frac{1}{4}(1+F)^2} + \frac{1-F}{(\Delta + V)^2 + \frac{1}{4}(1-F)^2}. \tag{60}$$

Comparing (59) with (60) illustrates the similarities mentioned in the introduction between the spectra of resonance fluorescence and spontaneous emission.

The intensity distribution for a greater number of atoms in the case of $k_0 R_{ij} \approx 1$ may be investigated numerically using equation (29) or (35). It is a function of the mutual

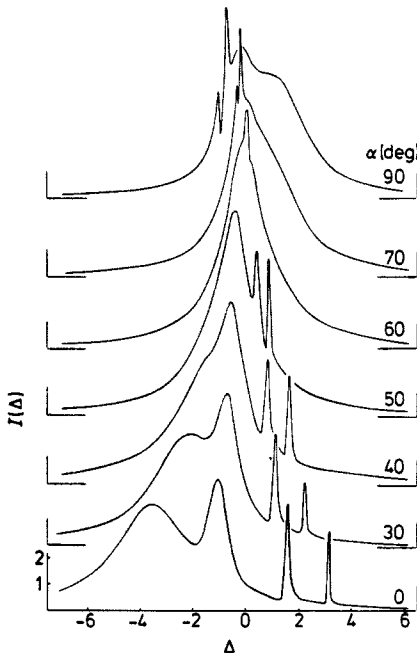


Figure 1. Intensity distribution of resonance fluorescence radiation emitted by four atoms placed on a straight line; $k_0 R_{12} = k_0 R_{13} = k_0 R_{23} = \rho = 1$ and α is the angle between the vector of the transition dipole moment (the same for all atoms) and the straight line defined by the atoms.

arrangement of atoms and the orientation of the transition dipole moment and as a rule is expressed by non-Lorentzian lines. To illustrate this case we present the spectra of four atoms placed on a straight line; $k_0 R_{12} = k_0 R_{13} = k_0 R_{23} \equiv \rho$ and α is the angle between the transition dipole moment \mathbf{d} of all the atoms and the straight line defined by the atoms. In figure 1 it is assumed $\rho = 1$ and α is variable. The set of spectra shows that with a fixed arrangement of atoms the change in the orientation of the dipole moment gives a change in the position of the spectral lines as well as in their halfwidths and heights. The number of lines in the spectra varies also. It is worth noting that the spectrum for $\alpha = 60^\circ$ has a shape similar to the Lorentzian spectrum of four atoms interacting with an electromagnetic field independently of each other. This effect is a consequence of the approximated equality

$$\mathbf{V} = \mathbf{0} \quad (61)$$

which is valid at $\alpha \approx 60^\circ$. It means that with the atomic positions and directions of dipole moments as above, the dispersion interaction between atoms is of small significance. Thus the change in the dipole moment orientation with a fixed atom arrangement may give a substantial change in the line shape. It is an effect similar to that mentioned above for a small sample.

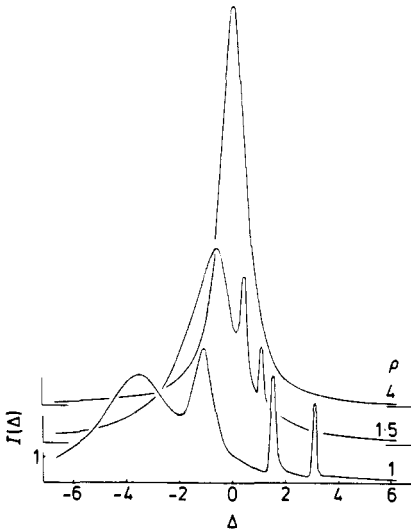


Figure 2. Intensity distribution for the same system as figure 1 with $\alpha = 0^\circ$.

In figure 2 $\alpha = 0^\circ$ is assumed and the parameter ρ is changed. Here the modifications of the spectra are caused by the changes in interatomic distances when the configuration of atoms and dipole moment orientations are kept constant. When ρ increases, the spectrum becomes identical with a Lorentzian spectrum of four isolated atoms (see (47)).

5. Conclusions

The emission spectrum of resonance fluorescence has been calculated for N two-level atoms with fixed positions. The basic assumption is that only one atom in the system

may be excited by the external electromagnetic field. It is valid when the intensity of the radiation scattered by the atoms is low. The calculations have been performed using the Heitler and Ma method. It is worth noting that the method is very useful for intensity distribution calculations because of its generality and mathematical simplicity and the ease of the physical interpretation of the results. The wider comparison of the Heitler and Ma method with others was given by Agarwal (1974). The real and virtual states in the expansion of the time-dependent state vector have been chosen to satisfy the above assumption and, moreover, they allow a solution of the problem without the rotating wave approximation (RWA). Doing without the RWA is an important element in the calculations since it ensures the proper form of cooperative energy shifts. Thus the approach of the present paper differs from others which use the typical Heitler and Ma method for problems of interaction between atoms and a weak field.

The intensity distribution derived (equation (29)) is expressed by two matrices \mathbf{F} and \mathbf{V} . Their elements describe the cooperative properties of the N -atom system interacting with field. The commutator of the matrices \mathbf{F} and \mathbf{V} qualifies an important feature of the spectrum, namely the possibility of representing the spectrum as a sum of Lorentzian lines. If matrices \mathbf{F} and \mathbf{V} commute then the spectrum is a sum of Lorentzian lines. The number of lines is equal to the number of atoms. The eigenvalues of \mathbf{F} and \mathbf{V} give halfwidth and shift of the lines, respectively. Each line is connected with the emission of radiation by one intermediate state. If \mathbf{F} and \mathbf{V} do not commute then the spectrum consists of N^2 components. Each of them is a positive defined rational function.

The results above may be applied to the determination of the properties of the spectrum of a weak electromagnetic field scattered by arbitrary two-level systems in relation to the number of systems, their spatial distribution and orientation of the transition dipole moments.

References

- Agarwal G S 1973 *Phys. Rev. A* **7** 1195–7
— 1974 *Springer Tracts in Modern Physics* vol 70 (Berlin: Springer)
Chang C S and Stehle P S 1971 *Phys. Rev. A* **4** 630–40
Czarnik J W and Fontana P R 1969 *J. Chem. Phys.* **50** 4071–4
Dicke R H 1954 *Phys. Rev.* **93** 99–111
Fontana P R and Hearn D D 1967 *Phys. Rev. Lett.* **19** 481–5
Freedhoff H S and van Kranendonk J 1967 *Can. J. Phys.* **45** 1833–59
Hearn D D and Fontana P R 1969 *J. Chem. Phys.* **51** 1871–80
Heitler W 1954 *The Quantum Theory of Radiation* (London: Oxford)
Heitler W and Ma S T 1949 *Proc. R. Ir. Acad.* **52** 109
Hutchinson D A and Hameka H F 1964 *J. Chem. Phys.* **41** 2006–11
Knight P L and Allen L 1973 *Phys. Rev. A* **7** 368–70
Lehmberg R H 1970 *Phys. Rev. A* **2** 889–96
Louisell W H 1964 *Radiation and Noise in Quantum Electronics* (New York: McGraw-Hill)
Milonni P W and Knight P L 1974 *Phys. Rev. A* **10** 1096–108
Saunders R and Bullough R K 1973 *J. Phys. A: Math., Nucl. Gen.* **6** 1360–74
Stephen M J 1964 *J. Chem. Phys.* **40** 669–73
Weisskopf V 1931 *Ann. Phys., Lpz.* **9** 23–66