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Abstract. When resonance fluorescence is excited in atoms whose excited states have Zeeman or hyperfine structure, it is to be expected that the character of the fluorescent light will be affected if the intensity of the exciting light is modulated or pulsed. An earlier theory of resonance fluorescence is developed to take account of such variations of the intensity of the exciting light. Resonance effects are predicted if the light is modulated at a frequency which coincides with a characteristic frequency of the atoms. A particular case, which has been studied experimentally, is worked out in detail. If the exciting light is pulsed, the prediction is that the fluorescent light will be modulated, in addition to being damped at the ordinary rate for spontaneous emission.

1. Introduction

Interest in resonance fluorescence has revived in recent years, partly because of the spectroscopic applications of 'double resonance' and 'level-crossing' experiments, and partly because of the illustrations which such experiments afford of the interactions between electromagnetic fields and free atoms.

As a natural development from earlier work, we have considered the consequences of exciting resonance fluorescence by light whose intensity is not constant in time; in particular, by modulated or pulsed light. By virtue of the time dependence of the intensity, a certain coherence is established between different spectral components of the light, so that when it is used to excite fluorescence in atoms whose excited states have Zeeman or hyperfine structure, the character of the fluorescent light is not the same as when the exciting light is uniform in time. If the exciting light is modulated, for example, the fluorescent light is itself modulated at the same frequency, but the amplitude of modulation undergoes changes which indicate an atomic resonance when the frequency of modulation coincides with the interval between two atomic energy levels.

Experiments using modulated light for the excitation of resonance fluorescence have been carried out, and are described in the accompanying paper (Corney and Series 1964). The excited states in question were the Zeeman components of the 5^3P_1 level of cadmium. Resonance phenomena were found when the frequency of modulation of the light coincided with the intervals between the Zeeman levels.

The object of this paper is primarily to derive detailed theoretical expressions for comparison with the experiments. For this purpose, it was convenient to extend a theoretical treatment of resonance fluorescence (Dodd and Series 1961, to be referred to as DS) which was developed to describe a double-resonance experiment of the type introduced by Brossel and Bitter (1952). The general expressions derived in § 2 of this paper, therefore, apply to experiments in which the atoms experience not only a static magnetic field and light whose intensity varies with time, but also a rotating magnetic

field. It is easy to separate the effects of the two time-dependent perturbations, so that the complication is not serious, but rather reveals interesting possibilities for further experiments. The simplification in which the rotating magnetic field is eliminated is made in § 3. In § 4 we show how the same results may be derived by the use of a classical model.

Since the earlier theoretical work was concerned with the Zeeman structure of the excited state, the formulae derived here do not apply explicitly to hyperfine or other types of structure. The case of a 2-level structure of unspecified origin would present no new difficulties, and more complicated cases could then be treated by methods of approximation. One would expect the general features of the results to be similar to the case of Zeeman structure treated here.

2. Extension of the theory of DS

The theory describes the behaviour of an atom in a uniform magnetic field $\mathbf{H}$ irradiated by light whose electric field at the atom at time t is $E(t)\mathbf{e}_i^0$, where $\mathbf{e}_i^0$ is a unit vector. The light is resonance radiation connecting the ground state $|g\rangle$ with a set of excited states $|m\rangle$ which belong to a given level of angular momentum $\mathbf{J}$. For simplicity, it is supposed that the value of J for the ground state is zero. (Particular examples are the resonance lines of zinc, cadmium and mercury, $^1P_1$ or $^3P_1$–$^1S_0$.) It was supposed in DS that the atoms were subjected also to a magnetic field $\mathbf{H}_1(t)$ rotating with angular frequency ω_0 in a plane perpendicular to $\mathbf{H}$. Although we are not now primarily concerned with the effects of $\mathbf{H}_1(t)$, we shall, for the time being, suppose that this field is present.

As in DS, we treat the process of excitation in detail and justify the approximations which are made. The resulting equation (6) could have been obtained more directly by use of the pulse approximation (Franken 1961) at the expense of insight into the nature of the problem.

We take up the argument of DS on p.360 in connection with the correlation integral

$$\langle \Phi(t_0, \tau) \rangle = \frac{1}{2T_0} \int_{t_0-T_0}^{t_0+T_0} E(t)E(t-\tau)\, dt \tag{1}$$

which we shall evaluate now explicitly as a function of t_0. We shall show that, for broad-band excitation, $\langle \Phi(t_0, \tau) \rangle$ has the same dependence on t_0 as $\langle \Phi(t_0, 0) \rangle$, which itself is proportional to the intensity of the light at time t_0. After the dependence on t_0 has been factorized out, the correlation integral may be treated as in DS. The rest of the work is straightforward. We take the opportunity of correcting two misprints in DS: on p.360, line 16, $E_i(t_0+\tau)$ should read $E_i(t_0-\tau)$; and on p.361, line 19, $\delta(k-k')$ should read $\delta(\tau)$.

The formal reduction of the correlation integral may be treated as follows. Define functions

$$\left.\begin{aligned} E(t_0, t) &= E(t) \\ E(t_0, t-\tau) &= E(t-\tau) \end{aligned}\right\} \text{ for } t_0+T_0 > t > t_0-T_0 \text{ and zero for all other values of } t.$$

The value of $\langle \Phi(t_0, \tau) \rangle$ is unaltered if these new functions are used in the integrand instead of $E(t)$ and $E(t-\tau)$. The new functions are introduced in order to allow the extension of the limits of integration from $t_0 \pm T_0$ to $\pm \infty$ without altering the value of the integral, thereby enabling it to be expressed in terms of the Fourier transform coefficients of $E(t_0, t)$ and $E(t_0, t-\tau)$. The two sets of Fourier coefficients are not, in fact, identical, but differ from each other by an amount which tends to zero as τ/T_0 tends to

zero. But, for quasi-monochromatic light of spectral width Δ, the correlation integral itself vanishes for values of $\tau \gg 1/\Delta$. If, therefore, we can ensure that $T_0 \gg 1/\Delta$, we may treat $E(t_0, t)$ and $E(t_0, t-\tau)$ as identical functions of the variable t, and we need not distinguish between the two sets of Fourier coefficients. Now, the upper limit for T_0 is set by the requirement $T_0 \ll 1/(\Gamma^2+x^2)^{1/2}$, so that the functions may be treated as identical provided $\Delta \gg (\Gamma^2+x^2)^{1/2}$. This condition is satisfied in the experiments with which we are concerned.

The Fourier coefficients $E(t_0, k)$ are defined through the equation

$$E(t_0, t) = \frac{1}{\sqrt{(2\pi)}} \int_{-\infty}^{+\infty} E(t_0, k) \exp(-ikt)\, dk. \tag{2}$$

Making use of the fact that $E(t_0, t-\tau)$ has the same Fourier coefficients as $E(t_0, t)$, we obtain

$$\langle \Phi(t_0, \tau) \rangle = \frac{1}{2T_0} \int_{-\infty}^{+\infty} |E(t_0, k)|^2 \exp(-ik\tau)\, dk. \tag{3}$$

We wish to relate this correlation function to the intensity of the incident light, i.e. to the mean value of $E^2(t_0)$, which is simply $\langle \Phi(t_0, 0) \rangle$. In the experiments which we wish to consider, the modulation was performed without altering the spectral distribution of the intensity, i.e. $\langle \Phi(t_0, 0) \rangle$ is independent of k. $|E(t_0, k)|^2$ may therefore be written $\sigma(t_0)\rho(k)$. It follows that

$$\langle \Phi(t_0, \tau) \rangle = \sigma(t_0) \int_{-\infty}^{+\infty} \rho(k) \exp(-ik\tau)\, dk. \tag{4}$$

We are not interested in the form of $\rho(k)$, though we have assumed that it extends smoothly over a sufficiently wide range of k.

We may now proceed as in DS to evaluate the integrals over τ for white light and for quasi-monochromatic light. The result is simply $\rho(k')$. (The case of monochromatic light may be treated by direct integration at an earlier stage.)

Returning to expression (26) of DS, we are left with the integral over t_0, which reduces to

$$\int_0^t dt_0 \sigma(t_0) \exp[-(\Gamma+ix)(t-t_0)]. \tag{5}$$

The general expression for the intensity corresponding to equation (32) of DS is now

$$I = \frac{4Nk_0^4}{\Gamma c^4 \hbar^2 r_0^2}\, \rho(k_0) \sum_{\substack{m,\mu,n \\ m',\mu',n'}} \mathscr{F}_{nn'} \langle m|\mu\rangle\langle\mu|n\rangle\langle n'|\mu'\rangle\langle\mu'|m'\rangle\, \mathscr{G}_{mm'}\Gamma$$
$$\times \exp[-i(m-m'-n+n')\omega_0 t] \int_0^t dt_0 \sigma(t_0) \exp[-(\Gamma+ix)(t-t_0)]. \tag{6}$$

2.1. *Modulated excitation*

We write $\sigma(t_0) = 1+\cos ft_0$. Provided $t \gg 1/\Gamma$, the integral over t_0 has the value

$$\frac{1}{\Gamma+ix} + \frac{\Gamma \cos ft}{(\Gamma+ix)^2+f^2} + \frac{f \sin ft}{(\Gamma+ix)^2+f^2}. \tag{7}$$

This result predicts that the fluorescent light will be modulated at the same frequency as the exciting radiation, as was to be expected. A more important prediction is that the amplitude of the modulation will show resonance phenomena when the applied frequency f is in the neighbourhood of any one of the frequencies x. These are the frequencies which are represented as differences between levels in the 'frequency diagram' (DS, p.366).

2.2. *Pulsed excitation*

We take $\sigma(t_0)$ equal to unity for a period of duration Θ centred on t_0, and zero elsewhere. The interesting phenomena are those which arise when Θ is much smaller than the lifetime of the atoms or the period of the frequencies x, whichever is the least. In such a case, the integral over t_0 has the value

$$\Theta \exp[-(\Gamma+ix)(t-t_0)]. \tag{8}$$

It is predicted that the fluorescent light will be modulated at the frequencies x, damped at the rate of spontaneous radiation from the atom. When $x = \Gamma$, the modulation will be critically damped.

3. Application to particular cases: static magnetic field

It has been convenient up to this point to retain the generality of a time-dependent magnetic field as well as time-dependent incident light. We now wish to isolate the effects which stem from the time dependence of the light. Removal of the rotating magnetic field is described by setting $H_1 = \omega_0 = 0$ in equation (25) of DS. (The static field **H** remains as before.) In this case, $\mu = n = m$, $\mu' = n' = m'$, and equation (6) reduces to

$$I = I_0 \sum_{m,m'} \mathscr{F}_{m,m'}\, \mathscr{G}_{m,m'}\, \Gamma \int_0^t dt_0 \sigma(t_0) \exp[-(\Gamma+ix)(t-t_0)] \tag{9}$$

where $x = (m-m')\omega$, $\omega = \gamma H$.

3.1. *The directions of the beams of light*

It is convenient to study the case in which the fluorescent light is taken at right angles to the direction of the incident light (unit vectors **j** and **i** respectively), and the field **H** is at right angles to both (unit vector **k**). The electric vector of the incident light makes an angle α_i with the direction of **H**; the fluorescent light is taken through an analyser which passes the electric vector inclined at the angle α to **H**. In this case the excitation and emission matrices for $J = 1$ are

$$\mathscr{F}_{m,m'} = |P|^2 \begin{pmatrix} \tfrac{1}{2}s_i^2 & \dfrac{-i}{\sqrt{2}}s_ic_i & \tfrac{1}{2}s_i^2 \\ \dfrac{i}{\sqrt{2}}s_ic_i & c_i^2 & \dfrac{i}{\sqrt{2}}s_ic_i \\ \tfrac{1}{2}s_i^2 & \dfrac{-i}{\sqrt{2}}s_ic_i & \tfrac{1}{2}s_i^2 \end{pmatrix} \quad \begin{matrix} \text{in which} & s_i = \sin\alpha_i \\ \text{and} & c_i = \cos\alpha_i \end{matrix} \tag{10}$$

and

$$\mathscr{G}_{m,m'} = |P|^2 \begin{pmatrix} \tfrac{1}{2}s^2 & \dfrac{-1}{\sqrt{2}}sc & -\tfrac{1}{2}s^2 \\ \dfrac{-1}{\sqrt{2}}sc & c^2 & \dfrac{1}{\sqrt{2}}sc \\ -\tfrac{1}{2}s^2 & \dfrac{1}{\sqrt{2}}sc & \tfrac{1}{2}s^2 \end{pmatrix} \quad \begin{matrix} \text{in which} & s = \sin\alpha \\ \text{and} & c = \cos\alpha \end{matrix}$$

3.1.1. *Modulated incident light.* Combining the expressions (7), (9) and (10), we have

$$\frac{I}{I_0\Gamma} = \cos^2\alpha_i \cos^2\alpha\left\{\frac{1}{\Gamma}+\cos ft\left(\frac{\Gamma}{\Gamma^2+f^2}\right)+\sin ft\left(\frac{f}{\Gamma^2+f^2}\right)\right\}$$
$$+\tfrac{1}{2}\sin 2\alpha_i \sin 2\alpha\left\{\left(\frac{\omega}{\Gamma^2+\omega^2}\right)+\tfrac{1}{2}\cos ft\left[\frac{\omega-f}{(\omega-f)^2+\Gamma^2}+\frac{\omega+f}{(\omega+f)^2+\Gamma^2}\right]\right.$$
$$\left.+\tfrac{1}{2}\sin ft\left[\frac{\Gamma}{(\omega-f)^2+\Gamma^2}-\frac{\Gamma}{(\omega+f)^2+\Gamma^2}\right]\right\}$$
$$+\tfrac{1}{2}\sin^2\alpha_i \sin^2\alpha\left\{\left[\frac{1}{\Gamma}-\frac{\Gamma}{\Gamma^2+4\omega^2}\right]+\tfrac{1}{2}\cos ft\left[\frac{2\Gamma}{f^2+\Gamma^2}-\frac{\Gamma}{(f+2\omega)^2+\Gamma^2}-\frac{\Gamma}{(f-2\omega)^2+\Gamma^2}\right]\right.$$
$$\left.+\tfrac{1}{2}\sin ft\left[\frac{2f}{f^2+\Gamma^2}-\frac{f+2\omega}{(f+2\omega)^2+\Gamma^2}-\frac{f-2\omega}{(f-2\omega)^2+\Gamma^2}\right]\right\}. \tag{11}$$

The result demonstrates that, with suitable orientations of polarizer and analyser, one may expect to find resonance effects in the amplitude of modulation, centred on the fields $H = f/\gamma$ and $f/2\gamma$, that is, when the applied frequency is equal, either to the Larmor precessional frequency of the excited atoms in the field H, or to twice that frequency. The effects may be interpreted as interferences between the σ and π, and between the σ^+ and σ^- Zeeman components of the fluorescent radiation. The coherence upon which the interference is based is generated by the coherence which the modulation imparts to the spectral components of the incident light.

Equation (11) is in good agreement with the experimental observations.†

3.1.2. *Pulsed incident light.* In this case, the expressions (8), (9) and (10) combine to give

$$\frac{I}{I_0\Gamma} = \Theta \exp[-\Gamma(t-t_0)]\{\cos^2\alpha_i \cos^2\alpha+\tfrac{1}{2}\sin 2\alpha_i \sin 2\alpha \sin\omega(t-t_0)$$
$$+\tfrac{1}{2}\sin^2\alpha_i \sin^2\alpha[1-\cos 2\omega(t-t_0)]\} \tag{12}$$

where Θ is the duration of the pulse applied at the time t_0.

In this case, the coherence which the pulsing imparts to the spectral components of the incident light is reflected in the modulation, which is a manifestation of interference between different Zeeman components of the fluorescent light.

Equation (12) has not yet been tested by experiment.

4. Classical model

In earlier papers (Dodd, Series and Taylor 1963, Kibble and Series 1963) it was shown that the results of certain modulation experiments could be interpreted by using a classical model of the fluorescing atoms. The present case may be treated similarly. The atoms are supposed to behave as isotropic oscillators which the exciting light sets into damped oscillation at their natural frequency. (This is the 'pulse approximation' which is based upon the assumption that the coherence time of the incident light is much smaller than the half-life of the oscillators.) The creation of dipoles is treated as a sequence of uncorrelated processes at a rate proportional to the intensity of the light.

† See note added in proof at end of article.

We treat the case when the geometrical arrangement is as specified in the last section. Consider the behaviour of one such dipole excited at time t_0. The component $P\cos\alpha_i$ of the amplitude **P**, parallel to the field **H**, will be unaffected by it. The component $P\sin\alpha_i$, perpendicular to the field, will, at time t, have precessed about it by the angle $\omega(t-t_0)$, where $\omega = \gamma H$. Both components will decay at the same rate, $\Gamma/2$. At time t, the component parallel to the analyser will be

$$P(t) = P[\cos\alpha_i\cos\alpha + \sin\alpha_i\sin\alpha\sin\omega(t-t_0)]\exp[-\tfrac{1}{2}\Gamma(t-t_0)].$$

To find the intensity of fluorescent light at time t, we multiply $|P(t)|^2$ by the number of dipoles excited in the interval dt_0, that is $\sigma(t_0)\,dt_0$, and integrate over t_0 from 0 to t.

The results are identical with those of the preceding sections.

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Note added in proof. Experiments have now been carried out in which atoms subjected to an oscillatory magnetic field as well as a static field were irradiated by modulated light. The results are in good agreement with expressions which are derived from equations (6), (7) and (10).