COHERENCE OF STATES IN THE SCATTERING OF MODULATED LIGHT

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Submitted to JETP editor January 31, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) **45**, 279-284 (August, 1963)

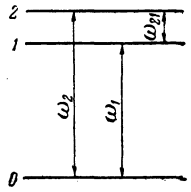
A theory of the scattering of intensity-modulated light is proposed for the case in which the atom of the scatterer has closely spaced excited states. The theory provides a description of experiments in which a resonance increase of the depth of modulation of the scattered light has been observed when the frequency of the modulation was equal to the Zeeman splitting of the excited term.

1. QUALITATIVE DISCUSSION

A recent paper by Aleksandrov[1] describes a very interesting experiment in which interference of closely spaced states of an atom is manifested. The light from a cadmium lamp ($^1S_0 \rightarrow {}^3P_1$ transition) was scattered by cadmium vapor which was in a weak magnetic field. The incident light was modulated in intensity owing to modulation of the power supply of the lamp, and was polarized in the plane perpendicular to the magnetic field. The magnetic field splits the upper term $^2P_1$ into a Zeeman triplet. The alternating component of the intensity of the light scattered at a fixed angle was observed as a function of the magnetic field. It was found that this component has a resonance maximum near the value of the magnetic field for which the frequency corresponding to the energy difference between the σ components of the triplet ($m = \pm 1$) is equal to the frequency of the modulation of the incident light. It must be emphasized that the Zeeman splitting is many orders of magnitude smaller than the Doppler width of the line.

This work is closely related to the experiments on the "crossing of levels," in which, with constant intensity of the incident light, it has been observed that there is a resonance change of the intensity of the scattered light as a function of the magnetic field.[2,3] Here the resonance is at the value of the magnetic field for which two Zeeman sublevels belonging to different excited states of the atom coincide.

In the present paper a theory of the scattering by an atom of light of variable intensity is proposed which takes into account the possibility of interference of the excited states. In particular this theory enables us to get a shape for the resonance curve which corresponds to Aleksandrov's experiment.



We first give a qualitative argument for the simplest example of a three-level system (see the figure). It is assumed for simplicity that the transitions $0 \rightarrow 1$ and $0 \rightarrow 2$ are electric dipole transitions, and that $\omega_{10}, \omega_{20} \gg \omega_{21}$. To each transition one can assign a corresponding oscillator with the respective natural frequencies ω_{10} and ω_{20} and the dampings $\gamma_1/2$ and $\gamma_2/2$. The scattering of light can be regarded as radiation from an oscillator executing forced vibrations under the action of the incident wave. The field of the incident wave is represented as a sum of harmonics. Then the motion of each oscillator is a superposition of harmonic vibrations, and the phase of each vibration is determined by the phase of the corresponding harmonic of the driving force.

Suppose that the incident light has a broad spectral composition, so that the spectral density remains practically unchanged over a range of frequencies of the order of ω_{21}. Each oscillator is intensely activated by those harmonics of the light which lie in a spectral interval of the order of γ near its resonance frequency.

Let us first consider the scattering of light whose intensity does not depend on the time (the experiment of the crossing of levels). If $\omega_{21} \gg \gamma_1 + \gamma_2$, then the oscillators 1 and 2 are excited by different harmonics of the incident light. The phases of the various harmonics are independent of each other (see below). After being averaged over the phases the square of the sum of the dipole moments of the two oscillators reduces to the sum

of the squares of the separate moments. Therefore the intensity of the scattered light will be equal to the sum of the intensities from the oscillators taken separately. If $\omega_{21} = 0$, then both oscillators are excited by the same harmonics, so that it is not the intensities, but the amplitudes for the scattered light that are added. If ω_{12} is of the order of $\gamma_1 + \gamma_2$, then some of the exciting harmonics will be common to the two oscillators; for these harmonics the amplitudes will add, and for the others, the intensities. Thus as ω_{12} is varied the intensity of the scattered light must show a resonance behavior near $\omega_{21} = 0$ with a resonance width of the order of $\gamma_1 + \gamma_2$.

Let us go on with the discussion of the scattering of light whose intensity varies harmonically with the time with frequency Ω. It will be shown below that in this case the phases of the harmonics ω and $\omega' = \omega \pm \Omega$ in the incident light are no longer independent. The variations of intensity of the incident light can be interpreted as beats of these correlated harmonics. Therefore the vibrations of the two oscillators will interfere not only when they are excited by the very same harmonics, but also when they are excited by two different harmonics with frequencies differing by Ω. The interference manifests itself in the occurrence of beats at the frequency Ω. That the amplitude of the beats will show a resonance behavior as a function of ω_{12} is obvious from the following arguments. The first oscillator is excited by harmonics in a band of frequencies near ω_{10} with width of the order of γ_1. The harmonics correlated with them are located in a shifted band at the frequency interval Ω from the first set of harmonics. The interference, which means the amplitude of the beats, will be largest when there is the most overlapping of the shifted band with the absorption band of the second oscillator.

Up to now we have not considered the vector properties of the dipoles corresponding to the transitions ω_{10} and ω_{20}. If, for example, these transitions are the σ components of a Zeeman triplet and the incident light is propagated along the magnetic field, then one is excited by a right-hand-rotation and the other by a left-hand component of the electric field vector. Interference of the light radiated by the two oscillators will occur only when the phases of the left-rotating and right-rotating components in the exciting light are correlated. There is such a correlation if the incident light is plane-polarized.

2. THE CORRELATIONS IN MODULATED LIGHT

Let us now establish the correlation properties of the light which were used in the foregoing discussion. We expand the electric field strength $\mathbf{E}(t)$ of the wave at a given point in a Fourier series

$$\mathbf{E}(t) = \sum_{\omega} \mathbf{E}_\omega e^{-i\omega t}, \quad \mathbf{E}_\omega = \frac{1}{T}\int_0^T e^{i\omega t}\mathbf{E}(t)\,dt. \tag{1}$$

For convenience the expansion is made for a large finite interval 0—T, so that the frequency values are $\omega = 2\pi n T^{-1}$ ($n = 0, \pm 1, \pm 2, \ldots$). The transition to infinite T and the corresponding Fourier integral can be made without difficulty by the substitution $T^{-1} = (2\pi)^{-1}d\omega$.

The correlation properties of the light are determined by the correlation function

$$S_{\mu\nu}(\omega, \omega') = \langle E^*_{\mu\omega}, E_{\nu\omega'}\rangle. \tag{2}$$

The angle brackets denote averaging over the ensemble of radiating systems. The indices μ and ν denote either rectangular or circular components of the field. If $S_{\mu\nu}(\omega, \omega')$ is not zero, this means that the phases of the corresponding harmonics are correlated. It is obvious that

$$S_{\mu\nu}(\omega, \omega') = T^{-2}\int_0^T d\tau \int_0^T d\tau' S_{\mu\nu}(\tau, \tau')\, e^{-i\omega\tau}e^{i\omega'\tau'},$$
$$S_{\mu\nu}(\tau, \tau') = \langle E_\mu(\tau), E_\nu(\tau')\rangle. \tag{3}$$

If the intensity of the light does not depend on the time, then $S_{\mu\nu}(\tau, \tau')$ depends only on the difference $\tau - \tau'$, so that

$$S_{\mu\nu}(\tau, \tau') = I\varphi_{\mu\nu}(\tau - \tau'), \tag{4}$$

where $I = E^2$ is proportional to the intensity of the light. In this case $S_{\mu\nu}(\omega, \omega')$ is different from zero only if $\omega = \omega'$:

$$S_{\mu\nu}(\omega, \omega)' = \delta_{\omega,\omega'} I\varphi_{\mu\nu}(\omega); \quad \varphi_{\mu\nu}(\omega) = T^{-1}\int_0^T e^{-i\omega\tau}\varphi_{\mu\nu}(\tau)\,d\tau. \tag{5}$$

The function $\varphi_{\mu\nu}(\omega)$ characterizes the spectral composition of the radiation. If the width of the spectral distribution is $\Delta\omega$, then the function $\varphi_{\mu\nu}(\tau - \tau')$ decreases rapidly when τ differs from τ' by more than about $(\Delta\omega)^{-1}$. If the intensity remains practically unchanged over a time of the order of $(\Delta\omega)^{-1}$ (in our case $\Delta\omega$ is the Doppler width of the line), then Eq. (4) can be generalized in an obvious way to the case of varying intensity [1]:

$$S_{\mu\nu}(\tau, \tau') = I(\tau)\,\varphi_{\mu\nu}(\tau - \tau') \tag{6}$$

[here it is quite immaterial whether we write $I(\tau)$ or $I(\tau')$].

[1] When the modulation of the light is caused by a modulation of the number of emitting atoms it can be shown that independently of the width of the spectrum $S_{\mu\nu}(\tau, \tau') = [\theta(\tau - \tau') \times I(\tau') + \theta(\tau' - \tau) I(\tau)]\varphi_{\mu\nu}(\tau - \tau')$. If $\Delta\omega \gg \Omega$, this is the same as Eq. (6).

For harmonically modulated light

$$I(t) = I_0(1 + \varepsilon \cos \Omega t) \tag{7}$$

and in this case, according to Eq. (3),

$$S_{\mu\nu}(\omega, \omega') = I_0 \varphi_{\mu\nu}(\omega')[\delta_{\omega,\omega'} + \tfrac{1}{2}\varepsilon(\delta_{\omega',\omega-\Omega} + \delta_{\omega',\omega+\Omega})]. \tag{8}$$

This formula indicates that there is a correlation between the phases of the harmonics ω and $\omega' = \omega \pm \Omega$.

Let us now turn to the correlation of the right-polarized and left-polarized components $E_+ = E_x + iE_y$ and $E_- = E_x - iE_y$. It is obvious that $S_{+-} = S_{xx} - S_{yy} - i(S_{xy} + S_{yx})$. For unpolarized light $S_{xx} = S_{yy}$ and $S_{xy} = S_{yx} = 0$, so that $S_{+-} = 0$. For light polarized along the x axis we have $S_{yy} = S_{xy} = S_{yx} = 0$, so that $S_{+-} = S_{xx} \neq 0$.

3. THEORY OF THE SCATTERING OF LIGHT OF VARYING INTENSITY

According to the correspondence principle the emitted field is determined by the dipole moment of the transition

$$d(t) = \int \psi_i^* \hat{d} \psi_0 \, d\tau.$$

Here ψ_i and ψ_0 are the respective wave functions of the initial and final states. If the initial state contains the excited states 1 and 2 with the amplitudes $c_1(t)$ and $c_2(t)$, then

$$d(t) = c_1^*(t) d_{10} + c_2^*(t) d_{20}.$$

The intensity $I_S(t)$ of the light scattered in a given direction can be expressed in terms of the density matrix $\hat{\rho}$ of the atom in the following way:

$$I_s(t) = K[\rho_{11}(t) \mid d_{10}^{\mu} \mid^2 + \rho_{22}(t) \mid d_{20}^{\mu} \mid^2 + 2 \operatorname{Re} \rho_{21}(t) d_{10}^{\mu} d_{02}^{\mu}]. \tag{9}$$

Here K is the ratio of the emitted intensity to the square of the dipole moment of the emitting atom. The quantity d^{μ} is the operator for the component of the dipole moment of the atom which is in the plane perpendicular to the direction of emission.

The Hamiltonian which determines the density matrix is

$$\hat{\mathcal{H}} = \hat{H} + \hat{V},$$

where $\hat{H}$ is the Hamiltonian of the atom and $\hat{V}$ is the interaction with the field of the incident wave. For simplicity we shall treat the field classically.[2)] We shall take into account the interaction with the zero-point oscillations, which leads to the finite lifetime, by adding an imaginary term to $\hat{H}$ ($\hat{H} = \hat{H}_0 - \frac{1}{2}i\hat{\Gamma}$; both $\hat{\Gamma}$ and $\hat{H}_0$ have only diagonal elements: $H_{mm} = \epsilon_m - i\gamma_m/2$).

The equation of motion of the density matrix is

$$i\hbar \dot{\rho}_{mn} = \sum_k (\mathcal{H}_{mk}\rho_{kn} - \rho_{mk}\mathcal{H}_{nk}^*).$$

In the first nonvanishing approximation in the perturbing field we have

$$i\hbar \dot{\rho}_{21} = \hbar(\omega_{21} - i\gamma_{21})\rho_{21} + V_{20}\rho_{01} - \rho_{20}V_{01}, \tag{10}$$

$$i\hbar \dot{\rho}_{20} = \hbar(\omega_{20} - i\gamma_{20})\rho_{20} + V_{20}, \tag{11}$$

where $\hbar\omega_{21} = \epsilon_2 - \epsilon_1$, $\gamma_{21} = \frac{1}{2}(\gamma_2 + \gamma_1)$. The equations for ρ_{12}, ρ_{11}, and ρ_{22} are analogous to Eq. (10), and those for ρ_{01}, ρ_{11}, and ρ_{22} to Eq. (11). In Eq. (11) we have set $\rho_{00} = 1$; V is the operator for the energy of interaction of the atom with the electric field E in the perturbing beam.

Let the incident light be propagated along the z axis and polarized in the plane XOZ. Then

$$V(t) = -\hat{d}^x E(t).$$

Substituting the solution of Eq. (11) (and of the analogous equation for ρ_{10}) in Eq. (10) and averaging over the ensemble of emitting systems, we get the following equation for ρ_{21}:

$$\dot{\rho}_{21} = -i(\omega_{21} - i\gamma_{21})\rho_{21} + F_{21}(t). \tag{12}$$

Here

$$F_{21}(t) = \hbar^{-2} d_{20}^x d_{01}^x \int_0^t dt' S(t, t')[e^{-i(\omega_{20} - i\gamma_{20})(t-t')} + e^{i(\omega_{10} + i\gamma_{10})(t-t')}], \tag{13}$$

where $S(t, t')$ is the correlation function defined by Eq. (3) (the indices μ, ν, which are equal to x, have been omitted).

We can use the expression (6) for S. Then, because of the sharpness of the function φ, we can [for $t \gg (\Delta\omega)^{-1}$] take the lower limit of the integral to be minus infinity. Also, for $\Delta\omega \gg \gamma$, ω_{21} we can set $\gamma_{10} = \gamma_{20} = 0$, $\omega_{20} = \omega_{10} = \omega_0$ in the exponents in Eq. (13). In this way we arrive at the following expression for F_{21}:

$$F_{21}(t) = \hbar^{-2} d_{20}^x d_{01}^x I(t) \varphi_{\omega_0}, \qquad \varphi_{\omega_0} = \int_{-\infty}^{\infty} e^{+i\omega_0 t} \varphi(t) \, dt. \tag{14}$$

Because $\varphi(t)$ is an even function the quantity $\varphi\omega_0$ is real.

Let us introduce the notations

$$d_{20}^x d_{01}^x = \tfrac{1}{2}(a_x + ib_x) \mid d_{01} \mid \mid d_{02} \mid, \qquad \rho_{21} = f_{21} + ig_{21}. \tag{15}$$

From Eq. (12) and Eqs. (14) and (15) we get the equation

[2)] The results are not changed by a quantum treatment of the electromagnetic field.

$$\ddot{f}_{21} + 2\gamma_{21}\dot{f}_{21} + (\omega_{21}^2 + \gamma_{21}^2) f_{21} = \tfrac{1}{2}[(\omega_{21}b_x + \gamma_{21}a_x) I(t) + a_x\dot{I}(t)]\hbar^{-2}\varphi_{\omega_0}|\mathbf{d}_{01}||\mathbf{d}_{02}| \quad (16)$$

and an analogous equation for g_{21} which is obtained from Eq. (16) by the substitutions $f_{21} \to g_{21}$, $a_x \rightleftarrows b_x$, $\omega_{21} \to -\omega_{21}$. If we write Eq. (9) in the form

$$I_s(t) = I_{11}(t) + I_{22}(t) + I_{21}(t) \quad (17)$$

and write $d_{10}^{\mu}d_{02}^{\mu} = \tfrac{1}{2}(a_\mu - ib_\mu)|\mathbf{d}_{01}||\mathbf{d}_{02}|$, then by using Eq. (16) we can get the following equations connecting the intensity of the scattered light with that of the incident light:

$$\ddot{I}_{21} + 2\gamma_{21}\dot{I}_{21} + (\omega_{21}^2 + \gamma_{21}^2) I_{21} = AI + B\dot{I}, \quad (18)$$

$$\dot{I}_{11} + \gamma_1 I_{11} = C_{11}I, \qquad \dot{I}_{22} + \gamma_2 I_{22} = C_{22}I. \quad (19)$$

Here

$$A = C_{21}[\omega_{21}(a_\mu b_x - a_x b_\mu) + \gamma_{21}(a_\mu a_x + b_\mu b_x)],$$
$$B = C_{21}[a_\mu a_x + b_\mu b_x],$$
$$C_{21} = 2|\mathbf{d}_{02}|^2|\mathbf{d}_{01}|^2\varphi_{\omega_0}(2\hbar)^{-2}K,$$
$$C_{nn} = |d_{0n}^{x}|^2|d_{0n}^{\mu}|^2\hbar^{-2}\varphi_{\omega_0}K. \quad (20)$$

If the transitions $1 \to 0$ and $2 \to 0$ are the σ components of a Zeeman triplet, the magnetic field is along the z axis, and the observation is made in a direction perpendicular to the field, then

$$\mathbf{d}_{20} = d(\mathbf{e}_x - i\mathbf{e}_y), \quad -\mathbf{d}_{10} = d(\mathbf{e}_x + i\mathbf{e}_y),$$
$$-d_{10}^{x} = d_{20}^{x} = d, \quad -d_{10}^{\mu} = d_{02}^{\mu} = de^{-i(\psi-\pi/2)},$$

where $\mathbf{e}_x$ and $\mathbf{e}_y$ are unit vectors along the axes in question and ψ is the angle between the direction of observation and the x axis. In this case

$$\tfrac{1}{2}C_{21} = C_{22} = C_{11} = C = K|\mathbf{d}_{01}|^4(2\hbar)^{-2}\varphi_{\omega_0},$$
$$A = -2C[\gamma_{21}\cos 2\psi + \omega_{21}\sin 2\psi], \qquad B = -2C\cos 2\psi,$$
$$\gamma_{21} = \gamma_2 = \gamma_1 = \gamma.$$

The formulas (18) and (19) describe the light scattered by one atom. To get the total intensity of the scattered light we must sum the solutions of Eqs. (18) and (19) over all the atoms of the scatterer, taking the Doppler effect into account. If $\omega_{21}v/c \ll \gamma$, then Eqs. (18) and (19) will also hold for the total intensity, and the summation changes only the constant C.

The quantity I_{21} is due to the interference of the excited states of the atom. Equation (18) enables us to interpret the effect observed by Aleksandrov as forced vibrations of the "oscillator" $I_{21}(t)$ under the action of the harmonic "driving force" $I(t)$. If $I(t) = I_0(1 + \epsilon\cos\Omega t)$, then

$$I_{21}(t) = AI_0/(\omega_{21}^2 + \gamma^2) - \varepsilon CI_0[(\omega_{21} - \Omega)^2 + \gamma^2]^{-1/2}\cos(\Omega t + \chi), \quad (21)$$

where $\chi = 2\psi - \tan^{-1}[(\Omega - \omega_{21})/\gamma]$.

In Eq. (21) it has been assumed that $\gamma \ll \omega_{21}, \Omega$. The first term, associated with the constant part of the intensity, describes the effect of "crossing of levels."

If the scatterer is irradiated with a pulse which is short in comparison with the lifetime of the atom, it can be seen from Eq. (18) that as the fluorescence dies out it will pulsate with the frequency ω_{21}. The possibility of such an effect has been pointed out in a paper by Podgoretskiĭ.[4]

The writers express their gratitude to E. B. Aleksandrov for acquainting them with his work before its publication. We are grateful to E. B. Aleksandrov and V. P. Kozlov for stimulating discussions.

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Translated by W. H. Furry
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