MODULATION OF SCATTERED LIGHT BY PARAMETRIC RESONANCE

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The resonance scattering of light on cadmium vapor in a magnetic field has been investigated theoretically and experimentally. The magnetic field, which splits the excited state into a Zeeman triplet, is modulated at a frequency Ω. The scattered light is found to be modulated at frequency Ω and harmonics of this frequency. The depth of modulation and the mean intensity of the luminescence exhibit resonance peaks when the frequency difference between the σ-components of the triplet is a multiple of Ω.

1. INTRODUCTION

AN experimental and theoretical description of the interference between two different energy states of an optically excited atom has been given in [1] and [2]. In that work the resonance scattering of plane-polarized light by cadmium vapor in a weak magnetic field was studied. The Zeeman splitting of the excited state was much smaller than the Doppler width of the line. It was found that the depth of modulation of the scattered light exhibits a resonance peak when the modulation frequency of the incident light is equal to the frequency difference between the σ-components of the Zeeman triplet.

In the present work we report on the theory and associated experiments concerning a related effect, the interference between two excited states produced by modulating the energy spacing between the states (cf. also [3] and [4]).[1)]

As in [1] and [2], we have studied the resonance scattering of light on cadmium vapor in a weak magnetic field. A diagram showing the levels in cadmium (even isotopes) is shown in Fig. 1. The number 2 denotes the state characterized by m = 1, the number 1 denotes the state characterized by m = − 1. The exciting light is plane-polarized and propagates along the magnetic field so that the m = 0 level is not excited and can be neglected in the analysis. In contrast with the conditions of the experiment reported in [1] and [2], in the present case the incident light is at a fixed intensity. However, the magnetic field that splits the excited state is modulated in such a way that the frequency difference between the two σ-components of the Zeeman triplet ω_{21} varies according to the relation

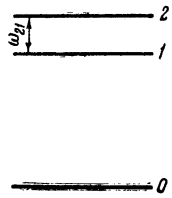

FIG. 1. Diagram of the 5^1S_0 and 5^3P_1 energy levels of cadmium in a magnetic field.

$$\omega_{21} = \omega_H + \omega_1 \sin \Omega t. \quad (1)$$

Under these conditions the intensity of the light scattered at a fixed angle is found to be modulated at frequency Ω and at multiples of this frequency. The depth of modulation and the mean intensity of the scattered light exhibit resonance peaks when $\omega_H = n\Omega$, where $n = 0, 1, 2, \ldots$, i.e., when the frequency of the transition between the levels $m = \pm 1$ is a multiple of the frequency at which the magnetic field is modulated.

In addition to the experiment on scattering of modulated light[1] and the experiment described here, modulated luminescence has also been observed in a double-resonance experiment[5]. In the latter case the variable magnetic field was perpendicular to the fixed field and produced transitions between sublevels of the Zeeman multiplet. In all three experiments the modulated luminescence was produced as a consequence of the formation of a coherent superposition of excited states in each atom of the scattering object; under these

[1)]A similar experiment has recently been carried out by means of modulated electron excitation. This work will be published in Optika i spektroskopiya (Optics and Spectroscopy).

conditions the phases of the excited states are the same for all atoms. These states have somewhat different energies and, consequently, radiate optical harmonics at different frequencies, which are capable of producing regular beats. From a formal point of view the criterion for coherent superposition of any two states a and b is the existence of a nonvanishing off-diagonal element of the density matrix ρ_{ab}.[6] As shown in our earlier work[2] the quantity ρ_{ab} can be interpreted as the displacement of some oscillator with a characteristic frequency $\omega_{ab} = \hbar^{-1}(\epsilon_a - \epsilon_b)$ and a damping equal to half the sum of the level widths of a and b. Forced oscillations of this oscillator are produced by the external periodic driving effect and the amplitude exhibits a resonance if the frequency of the driving force is close to the characteristic frequency ω_{ab}. In double-resonance experiments this driving force is the transverse variable magnetic field in combination with the fixed optical excitation. In scattering of modulated light the coherent superposition of states with $m = \pm 1$ arises because the light is plane-polarized. The excitation of the oscillator ρ_{ab} is due to the periodically varying intensity.

In the experiment described here the characteristic frequency of the oscillator ρ_{21} is varied periodically and a parametric resonance is produced. A qualitative interpretation can also be given in terms of two-quantum transitions, in the spirit of the work reported by Aleksandrov.[1] The coherence of the $m = \pm 1$ states is due to the fact that they can be excited by the same harmonic of the incident light; for example, if the $m = -1$ level is excited directly by a given harmonic the $m = +1$ level is also excited by a radiofrequency harmonic. We wish to emphasize that in itself the radio-frequency field in the present experiment does not cause transitions between sublevels of the excited state. The plane polarization of the exciting light provides complete correlation of the phases of the left and right-handed components of the light contributing to the excitation of the $m = \pm 1$ states.

All three of the effects described above can be used to determine the magnetic moment and the lifetime of the excited state. It will be shown below that the parametric resonance and scattering of the modulated light exhibit the feature that the position and width of the resonance curve are not affected by increasing the variable field (in this sense there is no saturation effect).

2. THEORY

The intensity of the light scattered by an atom having two nearby excited states (Fig. 1) is related to the density matrix of the atom as follows:[2]

$$I_s(t) = K\,[\rho_{11}|d_{10}^{\mu}|^2 + \rho_{22}|d_{02}^{\mu}|^2 + 2\,\mathrm{Re}\,\rho_{21}d_{10}^{\mu}d_{02}^{\mu}]. \quad (2)$$

Here, K is a factor that relates the intensity of the dipole radiation with the square of the dipole moment of the radiator; d^{μ} is the operator that gives projection of the dipole moment on a plane perpendicular to the direction of observation. The density matrix ρ is given by the following expressions:[2]

$$\dot{\rho}_{11} = -\gamma\rho_{11} + F_{11}, \qquad \dot{\rho}_{22} = -\gamma\rho_{22} + F_{22}, \quad (3)$$

$$\dot{\rho}_{21} + i\omega_{21}\rho_{21} = -\gamma\rho_{21} + F_{21}. \quad (4)$$

Here we assume that the widths of levels 1 and 2 are the same and equal to γ. If the incident wave propagates along the magnetic field and is polarized in the XOZ plane the quantity F is given by

$$F_{mn} = N\hbar^{-2}d_{m0}^{x}d_{0n}^{x}\,\langle\varphi_{\omega_0}\rangle\, I, \quad (5)$$

where $I = \langle E^2 \rangle$ is proportional to the intensity of the incident light (E is the electric field of the wave), $I\varphi_{\omega_0}$ is the spectral density of the incident light at frequency $\omega_0 = \omega_2 \approx \omega_1$ (it is assumed that the width of the spectral distribution of the incident light $\Delta\omega \gg \omega_{21}$), N is the number of scattering atoms. The angle brackets mean averages over the Doppler distribution of scatterer frequencies ω_0. The solution of Eq. (4) with the initial condition $\rho_{21}(0) = 0$ is

$$\rho_{21}(t) = F_{21}\int_0^t e^{-\gamma(t-t')}\exp\left\{-i\int_{t'}^{t}\omega_{21}(\tau')\,d\tau'\right\}dt'. \quad (6)$$

We introduce a new variable of integration $t' = t - \tau$. Then, at large values of the time $\gamma t \gg 1$ (for which transient effects disappear) the expression for $\rho_{21}(t)$ becomes

$$\rho_{21}(t) = F_{21}\int_0^\infty e^{-\gamma\tau}\exp\left\{-i\int_{t-\tau}^{t}\omega_{21}(\tau')\,d\tau'\right\}d\tau. \quad (7)$$

In the more or less trivial case where $\Omega \ll \gamma$ we can write

$$\int_{t-\tau}^{t}\omega_{21}(\tau')\,d\tau' \approx \omega_{21}(t)\,\tau,$$

and thus obtain

$$\rho_{21}(t) = F_{21}\,[\gamma + i\omega_{21}(t)]^{-1}. \quad (8)$$

Here, the change in $\rho_{21}(t)$ is a simple consequence of the intersection of levels with an adiabatic change in $\omega_{21}(t)$.

In the general case Eq. (7) can be written

$$\rho_{21}(t) = F_{21}\int_0^\infty d\tau\, e^{-(i\omega_H+\gamma)\tau}$$

$$\times\exp\left[-\frac{2i\omega_1}{\Omega}\sin\frac{\Omega}{2}\tau\,\sin\frac{\Omega}{2}(2t-\tau)\right]. \quad (9)$$

We expand the second exponential in the integral in Eq. (9) in Bessel functions, making use of the relation

$$e^{-iz\sin\varphi}=\sum_{k=-\infty}^{\infty}(-1)^k e^{ik\varphi}J_k(z). \qquad (10)$$

We then obtain the Fourier expansion

$$\rho_{21}(t)=F_{21}\sum_{k=-\infty}^{\infty}B_k e^{ik\Omega t}, \qquad (11)$$

where

$$B_k=(-1)^k\int_0^\infty d\tau\, e^{-(i\omega_H+\gamma)\tau}e^{-ik\Omega\tau/2}J_k\left(\frac{2\omega_1}{\Omega}\sin\frac{\Omega}{2}\tau\right). \qquad (12)$$

The Fourier coefficients B_k are obtained from the well-known formula

$$J_k\left(2a\sin\frac{\gamma}{2}\right)=e^{ik(\gamma-\pi)/2}\sum_{n=-\infty}^{\infty}J_{k+n}(a)\,J_n(a)\,e^{in\gamma},$$

which yields the following convenient expression

$$B_k=i^k\sum_{n=-\infty}^{\infty}J_{k+n}\left(\frac{\omega_1}{\Omega}\right)J_n\left(\frac{\omega_1}{\Omega}\right)[\gamma+i(\omega_H-n\Omega)]^{-1}. \qquad (13)$$

It is evident that all harmonics of the scattered light have resonance peaks when Ω is a multiple of the modulation frequency Ω.

We now rewrite Eq. (2), which gives the intensity of the scattered light, in the form

$$I_s(t)=I_1+I_2+I_{21}. \qquad (14)$$

Then, using Eqs. (3), (5), and (11) we have

$$I_1=I_2=CI/\gamma, \qquad (15)$$

$$I_{21}=-CI2\,\mathrm{Re}\left\{e^{2i\psi}\sum_{k=-\infty}^{\infty}B_k e^{ik\Omega t}\right\}. \qquad (16)$$

Here, the B_k are determined by Eq. (13); ψ is the angle between the direction of observation and the X axis; $C=NK|d_{01}|^4\langle\varphi\omega_0\rangle(2\hbar)^{-2}$. It is assumed that the line of observation is at right angles to the magnetic field. The interference term I_{21} exhibits a resonance when the spacing between the levels ω_H is a multiple of the modulation frequency of the magnetic field Ω. This term contains varying components at the frequency Ω and its harmonics.

We consider in greater detail the dc component and the first harmonic in I_{21}. We will assume that $\Omega\gg\gamma$ and that $\omega_H,\Omega>0$. In each case we consider only the resonance terms. We start with Eqs. (13) and (16).

1. The dc component $(k=0)$

$$I_{21}^{(0)}=-2CI\sum_{n=0}^{\infty}J_n^2\left(\frac{\omega_1}{\Omega}\right)[(\omega_H-n\Omega)^2+\gamma^2]^{-1/2}\cos(-2\psi+\chi_n)$$

$$=-2CI\sum_{n=0}^{\infty}J_n^2\left(\frac{\omega_1}{\Omega}\right)[\gamma^2+(\omega_H-n\Omega)^2]^{-1}[\gamma\cos 2\psi+(\omega_H-n\Omega)\sin 2\psi], \qquad (17)$$

where

$$\chi_n=\mathrm{arc\,tg}\frac{\omega_H-n\Omega}{\gamma}. \qquad (18)^*$$

It is evident that the dc component exhibits resonances when $\omega_H=0,\ \Omega,\ 2\Omega$ and so on. The resonance at $\omega_H=0$ $(n=0)$ with $\omega_1/\Omega\ll 1$ describes the well-known effect of level crossing.[7] The strength of the dc component of the scattered light at the resonances depends on the angle ψ between the direction of observation and the plane of polarization of the incident light.

2. The first harmonic $(k=+1,-1)$—modulation of the light at the field modulation frequency

$$I_{21}^{(1)}=-CI\,2\,\mathrm{Re}\,e^{2i\psi}(B_1e^{i\Omega t}+B_{-1}e^{-i\Omega t}).$$

a) The resonance at $\omega_H=0$ $(n=0)$, i.e., zero magnetic field

$$I_{21}^{(1)}=-4CIJ_1\left(\frac{\omega_1}{\Omega}\right)J_0\left(\frac{\omega_1}{\Omega}\right)\frac{-\gamma\sin 2\psi+\omega_H\cos 2\psi}{\gamma^2+\omega_H^2}\cos\Omega t$$

$$=-4CIJ_1\left(\frac{\omega_1}{\Omega}\right)J_0\left(\frac{\omega_1}{\Omega}\right)\frac{\sin(-2\psi+\chi_0)}{(\gamma^2+\omega_H^2)^{1/2}}\cos\Omega t. \qquad (19)$$

In this case the amplitude of the first harmonic of the scattered light depends on the angle ψ.

b) Resonances with $\omega_H=\Omega,\ 2\Omega$ etc.,

$$I_{21}^{(1)}=2CI\sum_{n=1}^{\infty}J_n\left[\frac{J_{n+1}^2+J_{n-1}^2+2J_{n+1}J_{n-1}\cos(-4\psi+2\chi_n)}{\gamma^2+(n\Omega-\omega_H)^2}\right]^{1/2}\times\sin(\Omega t+\Phi_n), \qquad (20)$$

where

$$\Phi_n=\mathrm{arc\,tg}\left[\frac{J_{n-1}-J_{n+1}}{J_{n-1}+J_{n+1}}\,\mathrm{tg}(-2\psi+\chi_n)\right].$$

The phase χ_n is determined by Eq. (18). The argument of the Bessel function is the quantity ω_1/Ω.

For weak modulation, in which case $\omega_1/\Omega\ll 1$,

$$I_{21}^{(1)}=2CI\sum_{n=1}^{\infty}\left(\frac{\omega_1}{2\Omega}\right)^{2n-1}\frac{1}{n!\,(n-1)!}\frac{\sin(\Omega t-2\psi+\chi_n)}{\sqrt{\gamma^2+(n\Omega-\omega_H)^2}}. \qquad (21)$$

In this case the amplitudes of the resonances in the scattered light are independent of ψ.

3. EXPERIMENT

1. Description of the apparatus. The experiment was carried out with Cd vapor at 200°C. We investigated the luminescence at $\lambda=3261$ Å (the $5^3P_1\to 5^1S_0$ transition). A general diagram of the apparatus is given in Fig. 2. The linearly polarized

*arc tg = $\tan^{-1}$; tg = tan.

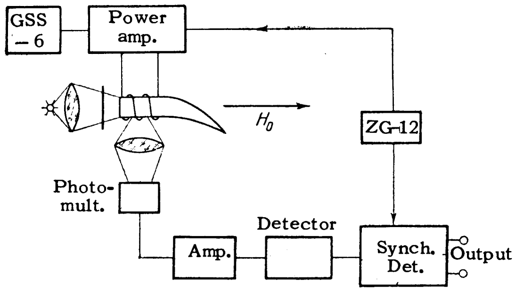

FIG. 2. Block diagram of the experimental apparatus.

light from an rf cadmium lamp excites the resonance luminescence in the Cd vapor in the container, which is a Wood's horn. The luminescence is detected at right angles by a photomultiplier (FÉU-46A). The signal from the photomultiplier is applied to a resonance heterodyne receiver, tuned to the frequency being observed. The amplified signal is then detected and applied to a synchronous detector and then to a recorder or voltmeter. The radio-frequency magnetic field is produced by a solenoid mounted on the horn. The solenoid is powered by a GSS-6 generator and a power amplifier. An additional modulation is applied in the latter (30 cps) to modulate the radio-frequency voltage applied to the solenoid. The 30-cycle modulation from a 3G-12 oscillator serves as a reference signal for the synchronous detector. A fixed magnetic field of the required strength and direction is produced by a system of Helmholtz coils.

2. Results. First harmonic. The modulation frequency of the field is fixed at 1030 kc/sec. The receiver is tuned to this frequency. In order to obtain the resonance curve the magnetic field which determines the splitting of ω_H is varied slowly. Resonances are observed when $\omega_H/\Omega = 0, 1, 2, 3, 4$ as predicted by Eqs. (19) and (20). The region of small depth of modulation of the magnetic field was investigated in detail.

At a low variable field ($\omega_1/\Omega \ll 1$) the strongest resonance occurs when $\omega_H = \Omega$. Equation (21), which gives the variable component of the scattered light, then becomes

$$I_{21}^{(1)} = CI \frac{\omega_1}{\Omega} \frac{\sin(\Omega t - 2\psi + \chi_1)}{\sqrt{\gamma^2 + (\Omega - \omega_H)^2}}. \qquad (22)$$

The resonance is expected at a field H = 0.245 Oe (the Landé factor is 1.5). In Fig. 3 we show a typical recording of the resonance curve. The time constant of the recorder is 3 sec. To compare the experimental line shape with the theoretical line shape we use Eq. (22) to compute $I_{21}^{(1)}$ for various points on the experimental curve. All the quantities obtained this way agree to within 5%. The mean value of γ is 4.1×10^5 sec^{-1}, corresponding to a lifetime $\tau = 2.4 \times 10^{-6}$. The latter value is in good agreement with the value of τ given for cadmium in the literature (cf. [8]). In analyzing the results account was taken of the nonlinearity of the detector at low signal amplitudes (approximately up to 10% of the level in Fig. 3).

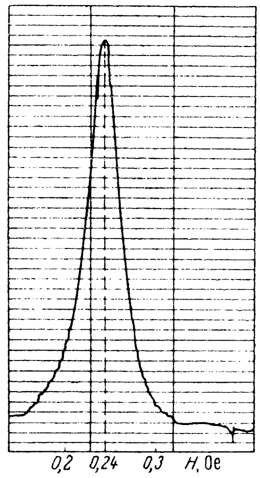

FIG. 3. Resonance line shape. The receiver is tuned to the field modulation frequency 1030 kc/sec.

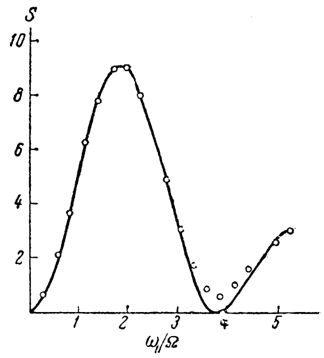

FIG. 4. The dc component of the radiation at resonance as a function of the depth of modulation of the field.

As predicted by the theory the intensity of the signal is independent of the angle between the direction of observation and the plane of polarization of the exciting light.

The linear behavior of the variable component of the intensity $I_{21}^{(1)}$ as a function of the amplitude of the variable field [Eq. (22)] was established taking account of the amplitude characteristics of the detector.

Resonances at $\omega_H/\Omega = 0, 2, 3, 4$ appear at relatively high amplitudes of the variable field. A careful investigation at high modulation values is hindered by two factors. The first is the need for careful shielding of the detector from the direct effect of the modulation voltage. The second is the excitation of atomic transitions between sublevels of the excited state caused by a spurious perpendicular component of the variable field. These transitions are especially strong when $\omega_H/\Omega = 2$, in which case the frequency Ω coincides with the frequency of the transition from the $m = \pm 1$ sublevels to the $m = 0$ sublevel (double resonance). This effect was established directly by observation of the polarization corresponding to the π-component in the scattered light.

3. Mean intensity (dc component). In this case the severe shielding requirements are relaxed and the experiment is simplified considerably; the signal from the photomultiplier is applied directly to the synchronous detector. The resonance at $\omega_H = \Omega$ was investigated in detail. The theory predicts a dependence of signal amplitude on ω_1/Ω corresponding to the square of the Bessel function of order one. This dependence is observed (Fig. 4). The solid curve is the function $J_1^2(\omega_1/\Omega)$ normalized to the signal peak. The experimental points are a good fit to the theoretical curve and a discrepancy is observed only when $\omega_1/\Omega \approx 4$. This discrepancy is explained by the residual double-resonance signal caused by the spurious transverse field component. The double-resonance peak lies at twice the value of the field H and is strongly broadened because of saturation.

The dc component exhibits one additional characteristic effect that has been discussed above, the dependence of the intensity on the angle between the plane of polarization of the light and the direction of observation; in this case we fix the value of the constant field (ω_H = const) and the depth of modulation. According to Eq. (17) the angular dependence should be of the form $I_{21}^{(0)} \sim \cos 2\psi$. In Fig. 5 this function is shown in polar coordinates together with the experimental points. A similar dependence is observed for the zero-field resonance at frequency Ω in accordance with Eq. (19).

4. The theoretical analysis shows that the general pattern of the effect is extremely complicated and diversified. In order to verify the theory we have undertaken to verify experimentally only certain specific features of the effect, in particular, those that distinguish it from double-resonance effects. In addition to observing the phenomena described above we have observed resonances at harmonics when the receiver was tuned to frequencies 2, 3, and 4 times the modulation frequency of the magnetic field.

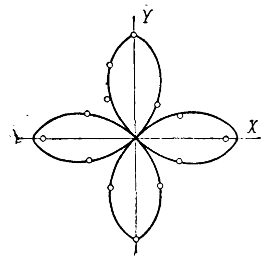

FIG. 5. Polar diagram of the signal magnitude corresponding to the dc component as a function of the angle between the direction of observation and the plane of polarization of the exciting light.

In conclusion we wish to indicate the attractive possibility of combining resonance scattering of modulated light with parametric resonances. In this case, when the field modulation frequency and the light modulation frequency are not the same the scattered light contains difference frequencies in addition to the fundamental and harmonics of the modulation frequency. This effect will be treated in a subsequent communication.

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