

INTERFERENCE EFFECTS IN RESONANT SCATTERING OF LIGHT BY ATOMS IN A MAGNETIC FIELD

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The angular and polarization distributions of resonantly scattered radiation in the case of intersection of the atomic levels are investigated by the method proposed in^[1,2]. The conditions for coherent excitation of these levels are studied as functions of the direction and polarization of the incident radiation. It is shown that: 1) linear and circular polarization of the scattered light are produced in the intersection region, 2) the scattering distribution pattern becomes anisotropic and is rotated with changing magnetic field intensity.

1. INTRODUCTION

In 1959, Colegrove, Franken, Lewis, and Sands proposed a spectroscopic method using interference effects in the intersection of resonant levels of atoms in a magnetic field^[1]. The main idea of the method is that at definite values of magnetic field intensity, corresponding to intersection of the coherently excited levels, there is observed a strong change of the angular and polarization distributions of the scattered light, owing to interference. This method makes it possible to use the observed dependence of the scattered-light intensity on the magnetic field intensity to determine certain characteristics of the atoms, namely the fine and hyperfine splitting, the Lamb shift, the radiative widths, and the magnetic-dipole and electric-quadrupole moments of the nucleus.

Subsequent papers^[2-8] gave a thorough calculation of the dependence of scattered-light intensity on the field intensity in the given observation direction, with allowance for the hyperfine splitting level repulsion, and optical pumping, and a comparison of this dependence with the corresponding experiments.

However, the influence of the intersection of the levels on the polarization of the scattered light was not investigated in these papers; furthermore, the conditions for coherent excitation of the levels were not sufficiently well analyzed.

We present here a calculation of the angular and polarization distributions of resonantly-scattered radiation upon intersection of the magnetic sublevels of the fine structure of the states $nP_{1/2}$ and $nP_{3/2}$, and ascertain the most favorable conditions for coherent excitation of the resonant levels and for organization of the appropriate experiments. The results are applicable to all atomic systems whose spectra are due to transitions of one S-valent electron ($nS - n'P - n''S$).

2. DISTRIBUTION OF THE PROBABILITY OF RESONANTLY-SCATTERED RADIATION

In the electric-dipole approximation, the amplitude of resonant scattering of light is^[9]

$$\langle k_2 e_2 \alpha_2 | S | k_1 e_1 \alpha_1 \rangle = \sum_{\alpha_m} \frac{\langle \alpha_2 | d e_2 | \alpha_m \rangle \langle \alpha_m | d e_1 | \alpha_1 \rangle}{(\omega - \omega_{m1}) + i\Gamma_m/2}, \quad (1)$$

where k_1 and e_1 are the wave vector and the polarization vector of the absorbed photon; k_2 and e_2 are the same vectors of the emitted photon; α_1 , α_m , and α_2 are the sets of quantum numbers characterizing the initial, intermediate, and final states of the atoms; $\hbar\Gamma_m$ is the radiative width of the resonant levels; $\hbar\omega_{m1} = \hbar(\omega_m - \omega_1)$ is the resonant energy, $\hbar\omega$ the absorbed-photon energy, and d the operator of the dipole moment of the atom.

Let us consider the stationary scattering of a radiation beam. The state of the photon is described in this case by the polarization density matrix. We assume that the radiation spectrum is constant within the limits of the group of resonant levels (the magnetic sublevels of the state $nP_{1/2}$ and $nP_{3/2}$, see below). This condition is realized as a result of the Doppler and collision broadenings of the lines in the resonant-radiation source. The state of the atom in the magnetic field at intensities $H \lesssim 10^4$ G and at the usual temperatures ($T \lesssim 10^3$ K) is described in terms of the populations of the magnetic sublevels of the ground state $nS_{1/2}$, and these populations can be regarded as equal with a high degree of accuracy, since $\mu_0 H/kT \ll 1$ and we are considering conditions under which optical pumping^[8] is immaterial.

From the amplitude (1) we can obtain an expression for the polarization density matrix of the radiation scattered in a given direction. To this end, we set up a bilinear quantity from (1), average in it over the initial states of the atom and over the polarization states of the initial radiation, and integrate over its spectrum. The basis of the polarization states is the representation of the circular polarization. The expression obtained in this manner contains cyclic components of the dipole-moment operator; these components are defined in a coordinate system connected with the wave vector and with the unit vectors of the polarization of the absorbed and emitted photons. The expressions also contains the polarization density matrix of the absorbed photon.

The symmetry axis of the atom is the magnetic field direction, so that it is advantageous to re-resolve the cyclic components of the dipole-moment operator in terms of their components in the coordinate system connected with the magnetic field. This transformation

is realized with the aid of the Wigner D-functions^[10]. The variable Ω contained in the function $D(\Omega)$ are the Euler angles (α, β, γ) defining the direction of the wave vector (α, β) and the orientation of the unit vectors of the linear polarization (γ) of the absorbed and emitted photons relative to the coordinate system connected with the magnetic field.

After performing the indicated transformations, we obtain the following expression for the polarization density matrix of resonantly scattered radiation, in the circular-polarization representation:

$$\langle \sigma | \mathcal{P}(\Omega_2) | \tilde{\sigma} \rangle = A \sum_{\alpha_m \alpha_n \alpha_{\nu} \alpha_{\mu}} (-)^{m+\mu} [D_{-\mu\sigma}^1(\Omega_2)]^* D_{-\mu\tilde{\sigma}}^1(\Omega_2) \times \frac{\langle \alpha_n | \tilde{d}_{\mu} | \alpha_n \rangle \langle \alpha_m | d_{\nu} | \alpha_1 \rangle \langle \nu | \rho(\Omega_1) | \tilde{\nu} \rangle \langle \alpha_2 | \tilde{d}_{\mu} | \alpha_n \rangle^* \langle \alpha_n | d_{\nu} | \alpha_1 \rangle^*}{(\omega_m - \omega_n) - i(\Gamma_m + \Gamma_n)/2}; \quad (2)$$

The summation is from $\sigma, \tilde{\sigma} = \pm 1$ —the indices of the circular polarization; $\Omega_{1,2}$ are the Euler angles for the absorbed and emitted radiation; d_{ν} and \tilde{d}_{μ} are the cyclic components of the dipole-moment operator;

$$\langle \nu | \rho(\Omega_1) | \tilde{\nu} \rangle = \sum_{\lambda, \tilde{\lambda}} D_{\lambda\tilde{\lambda}}^1(\Omega_1) \langle \lambda | \rho | \tilde{\lambda} \rangle [D_{\tilde{\lambda}\lambda}^1(\Omega_1)]^* \quad (3)$$

is the density matrix determined by the angular and polarization properties of the incident radiation; $\langle \lambda | \rho | \tilde{\lambda} \rangle$ is the polarization density matrix of the incident radiation in the circular-polarization representation $(\lambda, \tilde{\lambda} = \pm 1)$. The constant factor A in (2) is determined by the normalization, and is of no importance in the study of the relative intensities.

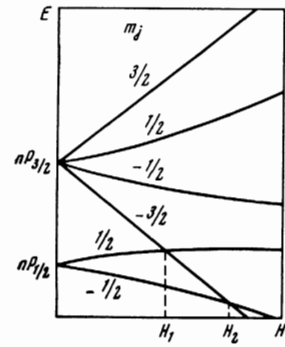
Formula (2) makes it possible to calculate the angular and polarization distributions of the scattered light. The terms of the sum (2) over the intermediate states α_m and α_n have different structures. The terms with $\alpha_m = \alpha_n$ do not depend on the frequency differences $\omega_m - \omega_n$ and are proportional to Γ_m^{-1} , whereas the terms with $\alpha_m \neq \alpha_n$ are proportional to the resonance factor $[\frac{1}{4}(\Gamma_m + \Gamma_n)^2 + \omega_m - \omega_n]^2$. The sum of such terms will be called the interference sum, since it contains different states that interfere in the case of coherent excitation.

3. COHERENCE CONDITIONS

The coherence of the states in the stationary case is determined, on the one hand, by the extent to which the corresponding energy levels are overlapped by the radiative widths, and on the other hand by the angular and polarization properties of the exciting radiation contained in the density matrix $\langle \nu | \rho(\Omega_1) | \tilde{\nu} \rangle$ (3). The figure shows the dependence of the energy of nP states of the fine structure on the magnetic-field intensity. As is seen from the figure, interference of the magnetic sublevels is possible in a "zero field," and also upon intersection of the magnetic sublevels $m_j = \pm \frac{1}{2}$ with $m_j = -\frac{3}{2}$ (the points H_1 and H_2).

On the other hand, analyzing the expression (2), we can easily find that the interference sum at the point H_1 is proportional to the matrix element $\langle \nu | \rho | \tilde{\nu} \rangle = \langle -1 | \rho | +1 \rangle$, and at the point H_2 to the matrix element $\langle \nu | \rho | \tilde{\nu} \rangle = \langle -1 | \rho | 0 \rangle$.

Let us calculate, in accordance with (3), the values of the density matrices $\langle \nu | \rho(\Omega_1) | \tilde{\nu} \rangle$ for a number of concrete cases of light scattering, with different states of the incident radiation.



a) Unpolarized light incident parallel to H . We have $\langle \pm 1 | \rho | \pm 1 \rangle = \frac{1}{2}$, and the remaining elements are equal to zero. We shall henceforth present only the nonzero matrix elements.

b) Unpolarized light incident perpendicular to H . We obtain $\langle \pm 1 | \rho | \pm 1 \rangle = \langle \pm 1 | \rho | \mp 1 \rangle = \frac{1}{4}$, $\langle 0 | \rho | 0 \rangle = \frac{1}{2}$.

c) Light linearly polarized in a direction parallel to H . We get $\langle 0 | \rho | 0 \rangle = 1$.

d) Light linearly polarized in a direction perpendicular to H . We obtain $\langle \pm 1 | \rho | \pm 1 \rangle = \langle \pm 1 | \rho | \mp 1 \rangle = \frac{1}{2}$.

e) Right-circularly polarized light incident parallel to H . We have $\langle +1 | \rho | +1 \rangle = 1$.

f) Right-circularly polarized light incident perpendicular to H . We obtain $\langle \pm 1 | \rho | \pm 1 \rangle = \langle \pm 1 | \rho | \mp 1 \rangle = \frac{1}{4}$, $\langle 0 | \rho | 0 \rangle = \frac{1}{2}$, $\langle \pm 1 | \rho | 0 \rangle = \sqrt{2}/4$.

We note that the foregoing limiting cases lead to conclusions concerning the intermediate cases of partly polarized radiation incident at angles other than zero and $\pi/2$ to the direction of H , since the angular dependence of $\langle \nu | \rho(\Omega_1) | \tilde{\nu} \rangle$ is smooth, and the partly polarized states are incoherent mixtures of the pure states considered above.

Thus, for coherent excitation of levels intersecting at the point H_1 , we can take radiation incident perpendicular to the direction of H (the cases b, d, and f). Particularly convenient in this respect is the case d, since it gives the relatively largest interference term.

For coherent excitation of the levels intersecting at the point H_2 , it is necessary to have radiation incident at an angle to H . The maximum effect is produced by case f. In addition, a detailed investigation shows that unpolarized radiation incident at an angle other than zero or $\pi/2$ to H , and linearly polarized radiation with a polarization vector direction within the same limits, give the required matrix element. The angle at which the maximum is reached is $\pi/4$.

Let us make some remarks concerning the description of the states of the atom in a magnetic field. Since we are considering the region of magnetic-field intensities at which the Zeeman splitting is of the order of the fine splitting, the spin I of the nucleus is no longer connected with the angular momentum (orbital plus spin) of the electron $j = 1 + s$ in the total angular momentum of the atom $F = I + j$, and the "good" quantum numbers are m_j, m_I , and I .

The quantum number j is "poor," since the magnetic splitting operator $V = \mu_0 H(g_L l + g_S s)$ ^[10] mixes the states with identical n, l , and m_j and with values of j differing by unity. The levels corresponding to

these states are repelled. In this case (see the figure) these are the magnetic sublevels with $m_j = \pm 1/2$ and $j = 1/2, 3/2$.

The "mixed" wave functions are determined by the expressions

$$\begin{aligned} |^{3/2}, m_j\rangle &= a |^{3/2}, m_j\rangle + b |^{1/2}, m_j\rangle, \\ |^{1/2}, m_j\rangle &= -b |^{3/2}, m_j\rangle + a |^{1/2}, m_j\rangle, \end{aligned} \quad (4)$$

where $|^{3/2}, m_j\rangle$ denotes a wave function that goes over at $\mu_0 H \ll \Delta E$ (the energy of the fine splitting at zero field) into the wave function with $j = 3/2$, $m_j = \pm 1/2$. We note that the states with $m = \pm 3/2$ do not mix.

The coefficients a and b are given in^[11]. Their values are

$$\begin{aligned} a &= \sqrt{1/2(1+\varepsilon)}, \quad b = \sqrt{1/2(1-\varepsilon)}, \\ \varepsilon &= \left(1 + \xi \frac{2m_j}{2l+1}\right) / \sqrt{1 + \xi \frac{4m_j}{2l+1} + \xi^2}, \quad \xi = \frac{\mu_0 H}{\Delta E}. \end{aligned}$$

Calculation of the parameter ξ at the intersection points H_1 and H_2 yields $\xi_1 = 2/9$ and $\xi_2 = 2/3$.

The dipole-moment operator matrix elements contained in (2) have the following form in the $|nlsjm_jIm_I\rangle$ scheme^[10]:

$$\begin{aligned} \langle n'l's_j'm_j'I'm_I' | d_\mu | nlsjm_jIm_I \rangle &= \delta_{II'} \delta_{m_I m_I'} (-)^{j'+l'+s_j-m_j'} \\ &\times ((2j+1)(2j'+1)(2l+1))^{1/2} \begin{pmatrix} j' & 1 & j \\ -m_j' & \mu & m_j \end{pmatrix} \begin{Bmatrix} l' & j' & 1/2 \\ j & l & 1 \end{Bmatrix} \langle n'l' \| d \| nl \rangle. \end{aligned} \quad (5)$$

We note that to calculate the angular and polarization distributions, it is not necessary to know $\langle n'l' \| d \| nl \rangle$, since all the spin-angle characteristics of the atom are separated in the $3j$ and $6j$ symbols and the factor preceding them in (5).

We shall henceforth assume that the instrument registering the scattered radiation does not resolve in the frequency the individual lines from the Zeeman components of the resonant levels. Then in formula (2) the summation is over all the magnetic sublevels of the initial ($nS_{1/2}$), intermediate (nP) and final levels ($nS_{1/2}$).

4. ANGULAR AND POLARIZATION DISTRIBUTIONS IN THE ABSENCE OF HYPERFINE SPLITTING

The interference of the magnetic sublevels $nP_{1/2}$ and $nP_{3/2}$ in a "zero field" and in the region of the points of intersection H_1 and H_2 (see the figure) leads to the appearance in the angular distribution of terms that depend on the azimuthal angle α , since in formula (2) the symbols μ and $\tilde{\mu}$ of the D functions are different ($D_{\mu\sigma}^j(\alpha, \beta, \gamma) = e^{-i\mu\alpha} D_{\mu\sigma}^j(0, \beta, 0) e^{-i\sigma\gamma}$). Thus, when the levels intersect, the axial symmetry is lost and the angular distribution becomes anisotropic in a plane perpendicular to the magnetic field.

In the case of interference of two magnetic sublevels with different m_j (points H_1 and H_2 in the figure), the coherent part of the angular distribution is given by

$$\begin{aligned} S_k &= B(\beta_1, \beta_2) (\Gamma_{mn} \cos \varphi_{12} - \omega_{mn} \sin \varphi_{12}) (\Gamma_{mn}^2 + \omega_{mn}^2)^{-1}; \\ \Gamma_{mn} &= (\Gamma_m + \Gamma_n) / 2, \quad \omega_{mn} = \omega_m - \omega_n, \quad \varphi_{12} = (\nu - \bar{\nu})(\alpha_2 - \alpha_1) \\ &= (\mu - \bar{\mu})(\alpha_1 - \alpha_2). \end{aligned}$$

This expression coincides with formula (10) of^[3]. It is convenient to rewrite it in the form

$$S_k(\alpha_1, \beta_1, \alpha_2, \beta_2) = B(\beta_1, \beta_2) R(H) \cos [\varphi_{12} + \delta(H)], \quad (6)$$

where $R(H) = [1 + \omega_{mn}(H)/\Gamma_{mn}]^{-1/2}$ is the resonant denominator and $\delta(H) = \tan^{-1}[\omega_{mn}(H)/\Gamma_{mn}]$ is the phase angle determining the direction of the symmetry axis of the angular distribution pattern in the plane perpendicular to H . Indeed, if we put in formula (6) $\beta_1 = \beta_1^0$, $\beta_2 = \beta_2^0$, $\alpha_1 = \alpha_1^0$, then the angular distribution will take the form $S_k(\alpha_2) \sim R(H) \cos [\Delta\nu(\alpha_2 - \alpha_1^0) + \delta(H)]$. Thus, in the region of interference of the magnetic sublevels we have $|\omega_{mn}(H)| \sim \Gamma_{mn}$; when the intensities change, the distribution pattern changes the degree of anisotropy and rotates at the same time. In the case of exact intersection $\omega_{mn}(H) = 0$, the degree of anisotropy is maximal and the distribution pattern is symmetrical with respect to the direction $\alpha_2 = \alpha_1^0$. The type of the pattern at the points H_1 and H_2 (see the figure) is different in accordance with the fact that for the point H_2 we have $|\Delta\nu| = 2$ and for the point H_1 we have $|\Delta\nu| = 1$.

We present numerical values of the angular distributions, calculated for the scattering cases b, d, and f and for the intensity regions in the vicinities of the points H_1 and H_2 . The angle $\alpha \equiv \alpha_2$ is reckoned in the cases b and f from the direction of \mathbf{k}_1 , and in the case d from the direction perpendicular to \mathbf{e}_1 ; the angle $\beta \equiv \beta_2$ is reckoned as before from the direction of H . For cases b and f in the region of H_1 we obtain $S(\alpha, \beta) = 1 + 0.17 \sin^2 \beta + 0.10 R(H) \sin^2 \beta \cdot \cos [2\alpha + \delta(H)]$. For the case d in the region of H_1 we get $S(\alpha, \beta) = 1 + 0.37 \cos^2 \beta + 0.20 R(H) \sin^2 \beta \cos [2\alpha + \delta(H)]$. In the case f in the region of H_2 we have $S(\alpha, \beta) = 1 + 0.25 \sin^2 \beta - 0.03 R(H) \sin 2\beta \cos [\alpha + \delta(H)]$. Thus, the interference "signal" in the region of H_2 is smaller by approximately one order of magnitude than in the region of H_1 . The optimal observation direction in the region of H_1 is $\beta = \pi(2k+1)/2$, and in the region of H_2 $\beta = \pi(2k+1)/4$. The "pure Lorentz" shape of the signal $S_k(H) \sim \Gamma_{mn}/(\Gamma_{mn}^2 + [\omega_{mn}(H)]^2)$ occurs in the region of H_1 at $\alpha = \pi k/2$ and in the region of H_2 at $\alpha = \pi k$.

All these conclusions agree with the results obtained in^[3,7]. In calculating the angular distributions it was assumed that the coefficients a and b (see (4)), which give a smooth dependence on the intensity, are constant in the intersection region and are equal to their values at the point of intersection. The relative accuracy of the results in this case is of the order of 10^{-2} in the region $|\omega_{mn}(H)| \sim \Gamma_{mn}$. The value of the ratio of the coherent part of the "signal" to the incoherent one is given in^[3] in the case of light scattering corresponding to our case b—unpolarized light incident perpendicular to H and scattered backwards (the region of H_1 is considered). This ratio is equal to 0.0895 at the maximum and agrees with our data within the limits of the calculation accuracy ($\sim 10^{-3}$).

The polarization of the scattered light changes in a resonant manner in the region of the level intersection. We present numerical values of the degrees of linear and circular polarization, calculated for the cases b, d, and f, in the regions of the intensities H_1 and H_2 at specified directions of the scattering angle $\beta \equiv \beta_2$ and of the position angle γ . It is reckoned in the same manner as the angle α when $\beta = 0$ and π , but is

reckoned from the direction of \mathbf{H} when $\beta = \pi/2$. We designate by η the degree of linear polarization and by \mathcal{P}^\pm the degree of right- and left-circular polarization (the upper and lower superscripts pertain to the scattering angles $\beta = 0$ and $\beta = \pi$, respectively).

In cases b and f, in the region of H_1 , at $\beta = 0$ and π and $\gamma = \pi/2 - (\frac{1}{2}) \tan^{-1} \delta(H)$ we have $\mathcal{P}^\pm = 0.07$, $\eta = 0.21R(H)$, and at $\beta = \pi/2$, $\gamma = 0$ we have $\mathcal{P}^\pm = 0$, $\eta = 0.14[1 - 0.71R(H) \cos[2\alpha + \delta(H)]]$.

In the case d, in the region of H_1 at $\beta = 0$ and π and $\gamma = \pi/2 - (\frac{1}{2}) \tan^{-1} \delta(H)$, we obtain $\mathcal{P}^\pm = -0.14$, $\eta = 0.27R(H)$, and when $\beta = \pi/2$, $\gamma = \pi/2$ we have $\mathcal{P} = 0$, $\eta = 0.38[1 + 0.32R(H) \cos[2\alpha + \delta(H)]]$.

In the case f in the region of H_2 , at $\beta = 0$ and π , and at $\gamma = \pi/2 - (\frac{1}{2}) \tan^{-1} \delta(H)$ we have $\mathcal{P}^\pm = 0.67$, $\eta = 0$, and when $\beta = \pi/2$, $\gamma = 0$, we have $\mathcal{P}^\pm = 0.03R(H) \times \cos[\alpha + \delta(H)]$, $\eta = 0.19$.

Thus, linear and circular polarizations, which do not exist far from the intersection point $|\omega_{mn}(H)| \gg \Gamma_{mn}$, appear in certain scattering directions.

5. INFLUENCE OF HYPERFINE SPLITTING

The analysis presented in the preceding section pertains directly to systems with nuclear spin $I = 0$, for example ${}^4\text{He II}$, ${}^{24}\text{Mg II}$, ${}^{26}\text{Mg II}$, ${}^{40}\text{Ca II}$, ${}^{28}\text{Si IV}$. In addition, the results of the section are applicable to systems having a hyperfine splitting, in the case when its magnitude is small compared with the energy width of the resonant level, for example in the case of HI, DI, and multiply-ionized atoms ($Z_a \gtrsim 3$). If the hyperfine splitting is large, then the dependence of the angular and polarization distributions on the field intensity will be more complicated (allowance for the hyperfine splitting is important for atoms of alkali metals and for their corresponding singly-ionized analogs).

To illustrate the influence of the hyperfine splitting on the angular and polarization characteristics of the scattered radiation at level intersections, it is convenient to consider the resonance curves—the plots of the intensities of the light scattered at given angles α_2 and β_2 against the magnetic field intensity. In the absence of hyperfine splitting, such a plot has a maximum and a minimum if $\cos \varphi_{12} = 0$ (see (6)), or one maximum (minimum) if $\sin \varphi_{12} = 0$. In the presence of hyperfine splitting, superposition of the resonance curves from different hyperfine components takes place upon intersection. Since the points of intersection are shifted relative to one another in intensity, the degree of this superposition depends on the ratio of the hyperfine splitting to the radiative width. It is important to note

that in the case of resonant transitions the values of I and m_I remain unchanged, so that in our case only the hyperfine components $m_j = \pm \frac{1}{2}$ and $m_j = -\frac{3}{2}$ with identical values of m_I interfere. Thus, there can be $2I + 1$ maxima or minima on the resonance curve.

In analyzing the hyperfine splitting in the region of intersection of the magnetic sublevels $m_j = -\frac{3}{2}$ and $m_j = -\frac{1}{2}$ (the point H_2), it is necessary to take into account the repulsion of the components that obey the selection rules $m'_j = m_j \pm 1$, $m'_I = m_I \mp 1$, since the Ij interaction mixes these states. This repulsion is accounted for in^[4-7] for HI, ${}^6\text{Li I}$, and ${}^7\text{Li I}$. There is no repulsion for the intersection of $m_j = +\frac{1}{2}$ and $m_j = -\frac{3}{2}$ (the point H_1), since $\Delta m_j = 2$.

In conclusion we note that the results obtained in this paper make it possible to modernize the method proposed in^[1-2]. Namely, information concerning the structure of the levels of the atoms can be obtained from observations of the dependence of the polarization of scattered light on the magnetic field intensity. New experimental possibilities are also uncovered by the obtained effect of rotation of the scattering distribution pattern..

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