The Zeeman Effect and Electronic Stimulated Raman Scattering

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The stimulated Raman scattering (SRS) susceptibility for atomic levels in a stationary magnetic field H is calculated by the density-matrix technique. The SRS amplification coefficient is represented by a series in powers of H. The separate terms of the series are estimated. Selection rules for SRS in a stationary magnetic field are obtained and it is shown that in this case intercombination transitions are allowed. Besides the opportunity this phenomenon offers for observation of such transitions, it is also interesting in that it permits one to vary the scattered-light frequency from zero to several tens of inverse centimeters by varying H from zero to 10^5 Oe.

A tunable laser based on stimulated Raman scattering (SRS) of infrared radiation in semiconductors in a magnetic field was recently developed. The change of frequency of the scattered quantum is determined in this case by the spacing between the Landau levels, which is varied by the magnetic field[1-4]. Insofar as we know, however, SRS on electronic levels of free atoms in a magnetic field H, tunable by means of a magnetic field, has never been investigated before. The present paper is devoted to this question.

The Zeeman effect lifts the degeneracy of the atomic levels with respect to the magnetic quantum number m, and each level splits into a series of magnetic sublevels, from which Raman scattering is possible. The distance between the magnetic sublevels varies with H, and the frequency ω_S of the scattered light varies with it:

$$\Delta\omega_s = \omega_s^{(0)} - \omega_s^{(H)} = \mu_B H(g_1 m_1 \pm g_2 m_2), \qquad (1)$$

where $\omega_S^{(0)}$ and $\omega_S^{(H)}$ are the frequencies of the scattered light without a field and in a field H, μ_B is the Bohr magneton, and g_1 and g_2 are the Lande factors of the initial and final levels, which are magnetic sublevels of different electronic levels of the atom. The plus sign is used in (1) if the signs of m_1 and m_2 differ, and the minus sign if they are the same. At $0 < H < 10^5$ Oe the value of $\Delta\omega_S$ for this system of levels changes from zero to several dozen reciprocal centimeters. This uncovers a possibility of smoothly tuning the frequency of the scattered light by continuously varying the magnetic field.

Let us calculate the susceptibility of the nonresonant SRS process on atoms in a constant magnetic field in the steady state, using the method of[6] to solve the density-matrix equations. We write down the Hamiltonian in the form $W = W_0 + V(t)$. The unperturbed Hamiltonian W_0 consists of the free-atom Hamiltonian and the energy of interaction with the external constant magnetic field, $\mu \cdot H$, where $\mu = (e\hbar/2mc)(L + 2S)$ is the magnetic dipole moment operator of the atom. The interaction Hamiltonian is equal to

$$V(t) = -\frac{e}{mc}\mathbf{pA} + \frac{e}{2mc^2}\mathbf{R}[\mathbf{AH}], \qquad (2)^*$$

where p and R are the operators of the momentum and of the electric dipole moment of the atom, and

$$\mathbf{A} = {}^1/_2[\mathbf{A}_L{}^0 \exp(i\omega_L t) + \mathbf{A}_S{}^0 \exp(-i\omega_S t) + \text{к.с.}] \qquad (3)$$

is the vector potential of the radiation field, consisting of laser and Stokes components.

The calculation was carried out for the three-dimensional case, so that we can consider the susceptibility for different polarizations of the laser and Stokes radiation and for different orientations of the field H and the wave vectors $\mathbf{k}_L$ and $\mathbf{k}_S$. We consider the case when $\mathbf{k}_L \parallel \mathbf{k}_S \parallel \mathbf{H}$ and the incident and scattered lights are linearly polarized. Solving the system of equations for the density-matrix elements ρ_{1l}, ρ_{l2}, and ρ_{12}, where 1, 2, and l are respectively the initial, final, and intermediate levels (l is an arbitrary number), we obtain the expressions for the density matrix elements, which are needed for the calculation or the nonlinear polarization at the frequency ω_S:

$$\rho_{1l} = \frac{1}{\hbar}\rho_{12}{}^+\left(-\frac{i\omega_{2l}}{2c}\mathbf{A}_L{}^{0*} + \frac{e}{4mc^2}[\mathbf{A}_L{}^{0*}\mathbf{H}]\right)\mathbf{R}_{2l}\frac{\exp(-i\omega_S t)}{\omega_{1l} - \omega_S},$$

$$\rho_{l2} = -\frac{1}{\hbar}\left(-\frac{i\omega_{1l}}{2c}\mathbf{A}_L{}^{0*} + \frac{e}{4mc^2}[\mathbf{A}_L{}^{0*}\mathbf{H}]\right)\mathbf{R}_{1l}\rho_{12}{}^+\frac{\exp(-i\omega_S t)}{\omega_{l2} - \omega_S},$$

$$\rho_{12}{}^+ = \frac{N_1\hbar^{-3}}{\Lambda - i\delta_{12}}\sum_l\left\{\frac{\mathbf{R}_{1l}}{\omega_{l1} - \omega_L}\left(-\frac{i\omega_{1l}}{2c}\mathbf{A}_L{}^0 + \frac{e}{4mc^2}[\mathbf{A}_L{}^0\mathbf{H}]\right)\right.$$
$$\times\mathbf{R}_{l2}\left(-\frac{i\omega_{l2}}{2c}\mathbf{A}_S{}^0 + \frac{e}{4mc^2}[\mathbf{A}_S{}^0\mathbf{H}]\right)$$
$$+\frac{\mathbf{R}_{1l}}{\omega_{l2} + \omega_L}\left(-\frac{i\omega_{1l}}{2c}\mathbf{A}_S{}^0 + \frac{e}{4mc^2}[\mathbf{A}_S{}^0\mathbf{H}]\right)$$
$$\left.\times R_{l2}\left(-\frac{i\omega_{l2}}{2c}\mathbf{A}_L{}^0 + \frac{e}{4mc^2}[\mathbf{A}_L{}^0\mathbf{H}]\right)\right\}; \qquad (4)$$

here N_1 is the population of the initial level, $\delta_{12} = \tau_{12}^{-1}$, where τ_{12} is the relaxation time for the $1 \to 2$ transition, and $\Lambda = \Delta + \Delta^*$, where $\Delta = \omega_L - \omega_S - \omega_{21}$.

If the incident and scattered lights are linearly polarized and the dispersion of the Faraday effect can be neglected, then the quantity $\operatorname{Re}\Delta^*$, which determines the frequency shift of the $1 \to 2$ transition, and therefore also the frequency shift of the Stokes radiation in the strong optical field, is given by

$$\operatorname{Re}\Delta^* = -\frac{1}{4\hbar^2}\left\{\frac{|E_L{}^0|^2}{\omega_L{}^2}\beta_{vv}^{(1)} + \frac{|E_S{}^0|^2}{\omega_S{}^2}\beta_{vv}^{(2)}\right\}, \qquad (5)$$

where E_L^0 and E_S^0 are respectively the electric field intensities of the laser and the Stokes waves. Assuming that $|E_L^0|^2 \gg |E_S^0|^2$, we can neglect the second term in the right-hand side of (5). Furthermore,

$$\beta_{vv}^{(1)} = \sum_l\left(\frac{\omega_{l2}{}^2 R_{l2v} R_{2lv}}{-\omega_L + \omega_{2l}} - \frac{\omega_{l1}{}^2 R_{1lv} R_{l1v}}{\omega_L + \omega_{1l}}\right). \qquad (6)$$

*$[\mathbf{AH}] \equiv \mathbf{A} \times \mathbf{H}$.

The imaginary part of Δ^* (the broadening of the $1 \rightarrow 2$ line), due to the presence of $i\tau^{-1}$ in the denominators of the exact expression for Δ^*, can be neglected in the considered case of nonresonant scattering.

From the expression for the polarization vector at the Stokes frequency[6,7]

$$\mathbf{P}(\omega_S) = \frac{e}{m} \int_{-\infty}^{t} \mathrm{Sp}\, \hat{\rho}(t', \omega_S) \hat{\mathbf{p}}\, dt' \qquad (7)$$

we obtain the susceptibility χ'' and then, using the known formula[8], the SRS gain

$$g_l = 4\pi\omega_S \chi'' / c. \qquad (8)$$

Expanding χ'' in powers of H, we can write, accurate to $O(H^2)$,

$$g_l = \frac{4\pi\omega_S}{c}(\chi_0'' + \chi_1''H + \chi_2''H^2) = g_{l0} + g_{l1} + g_{l2}.$$

After calculating χ_0'', χ_1'', and χ_2'' under the assumption that the wave functions in the matrix elements are independent of H, but only the frequencies are field-dependent, as is the case of the simple Zeeman effect, we obtain the relations

$$g_{l0} : g_{l1} : g_{l2} = \hbar\omega_L : \mu_B H : \mu_B^2 H^2 / \hbar\omega_L.$$

This relation is approximate and is valid if the frequencies of the laser and Stokes light and the frequency of the $1 \rightarrow 2$ transition are of the same order. At $\omega_L \sim 10^{14}$ sec^{-1} and $H \sim 10^5$ Oe, this ratio is $10^4 : 10^2 : 1$.

To estimate the absolute value of χ_0'', we consider by way of an example the transition $6^3P_1(m = -1) \rightarrow 6^3P_2(m = +1)$ in the mercury atom. The state 6^3P_1, which is located at a distance 39 409 cm^{-1} from the ground state $^1S_0$, can apparently be populated by using as the pump the four harmonic of a glass: Yb^{3+} laser, whose frequency is 39 409 cm^{-1} [9]. One magnetic sublevel $k_1 = -1$ can be populated by pumping with left-polarized light. The source of the SRS can be lasing in Pb at 24 643.7 cm^{-1} (λ = 4057.83 Å)[10]. An analysis of the Hg spectrum[11] shows that in this case only the state 7^3S_1 plays any role (the level scheme is shown in the figure), and then

$$\chi_0'' = -\frac{N_1 |E_L^0|^2 \delta_{12} \omega_{l1}^{(0)2} \omega_{2l}^{(0)2}}{8\hbar^3 (\Lambda^2 + \delta_{12}^2)\, \omega_L^2 \omega_S^2} |R_{\nu l1} R_{\nu 2l}|^2 \left(\frac{1}{\omega_{l2}^{(0)} + \omega_L} + \frac{1}{\omega_{l1}^{(0)} - \omega_L} \right)^2,$$

where l should be taken to mean $^3S_1$.

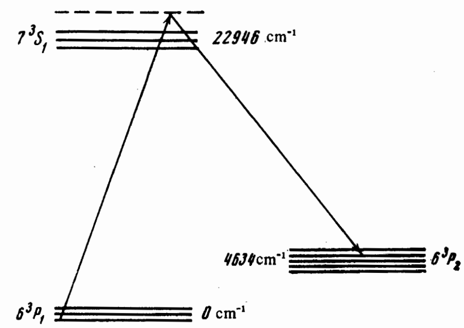
Using the connection between the oscillator strength and the square of the electric dipole moment[12], and the values of the oscillator strengths of the transitions of interest to us[11], we get

$$|R_{\nu l1}|^2 \approx 2.2 \cdot 10^{-35} \text{ g-cm}^5\text{sec}^{-2}, \quad |R_{\nu 2l}|^2 \approx 1.8 \cdot 10^{-35} \text{ g-cm}^5\text{sec}^{-2}.$$

Substituting in (5) and (6) these quantities, the aforementioned frequency ω_L = 24 643.7 cm^{-1}, and the transition frequencies indicated in the figure, we obtain

$$\beta_{\nu\nu'}^{(1)} \approx 10^{-19} \text{ g-cm}^5\text{sec}^{-3} \quad \mathrm{Re}\,\Delta^* \approx 10^{-6} |E_L^0|^2 \text{ sec}^{-3}$$

To estimate the order of magnitude of χ_0'', we put $N_1 = 10^{17}$ cm^{-3}, $|E_L^0|^2 \approx 10^7$ g-cm^{-1}sec^{-2}, $\Lambda = \delta_{12} \approx 1$ cm^{-1}. We then have $\omega_S = \omega_L - \omega_{21} + \Delta^* - \Lambda \approx 20\,000$ cm^{-1} and a final estimate $|\chi_0''| \approx 10^{-4}$. From[8] we obtain a gain $g_{l0} \approx 50$ cm^{-1}. According to formula (1), the change of frequency $\Delta\omega_S$ for the transition in question is 80 cm^{-1} at $H = 10^5$ Oe.



To obtain the transformation properties of the susceptibility tensor and to derive the selection rules, we note that χ_0'', χ_1'', and χ_2'' consist of products of tensors of different ranks, from which irreducible spherical tensors can be made up[13,14]. In particular, χ_2'' is a product of two third-rank tensors whose transformation properties are governed by terms containing products of two components of the matrix elements of the electric dipole moment and one component of the magnetic dipole moment. They can be used to form the irreducible third-rank tensors T_{30} and $T_{3\pm 2}$. The spin component μ in χ_2'' gives rise to the presence of bilateral tensor operators $T_{\omega r}$[14], the matrix elements of which are expressed in terms of 9_j symbols in the case of LS coupling:

$$\begin{pmatrix} J & S & L \\ \omega & q & p \\ J' & S' & L' \end{pmatrix}$$

in which J is the quantum number of the total angular momentum, ω the rank of the bilateral tensor, p is its rank with respect to the spatial variables, and q is its rank with respect to the spin variables. The 9_j symbol differs from zero if the vectors in the rows and in the columns satisfy the triangle rule[14], from which it follows that

$$|\Delta J| = \omega,\ \omega - 1, \ldots 0; \ |\Delta L| = p,\ p - 1, \ldots 0; |\Delta S| = q,\ q - 1, \ldots 0.$$

For Δm we have the following selection rule[12-14]: $\Delta m = 5$. In addition, owing to the parity selection rule, $|\Delta L|$ is an even number. Since in our case $\omega = 3$ and $r = 0, \pm 2$, we obtain

$$|\Delta J| = 3, 2, 1, 0; \quad |\Delta L| = 2, 0; \quad |\Delta S| = 1, 0; \quad \Delta m = 0, \pm 2.$$

These selection rules differ from those obtained by Placzek[15] for electronic Raman scattering; this makes it possible to obtain, in principle, new transitions and to determine the positions of atomic levels.

In addition to this conclusion, we can make the following deduction concerning the possible applications of smooth tuning of the frequency of scattered light with the aid of a magnetic field. First, the presently existing tunable lasers operate in the infrared and in the visible bands[1]; atomic transitions can have frequencies also in the ultraviolet, so that a possibility of tuning in the ultraviolet is uncovered. Second the possibility of frequency tuning in weak magnetic fields, within the limits of the line width of an atomic transition, will make it possible, in principle, to use radiation scattered in a field H as a source of exitation of resonant SRS for the study of the frequency dependence of its parameters.

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