Physics Letters A 169 (1992) 463–468
North-Holland

PHYSICS LETTERS A

Dynamic Stark effect action on optical pumping of atoms in an external magnetic field

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Received 10 June 1992; accepted for publication 17 August 1992
Communicated by B. Fricke

The influence of the dynamic Stark effect on the optical pumping of atoms in a magnetic field, using the broad band approximation, is examined. It is demonstrated that the dynamic Stark effect can lead to a nonlinear effect on the light intensity conversion of alignment produced by linearly polarized light in the orientation of the angular momentum of atoms.

Practically immediately after the appearance of laser sources they became widely employed for optical pumping of atoms, see for example ref. [1]. The interaction between the laser radiation and the atoms must be of resonant nature. For this reason one and the same kind of atoms were at first used both for laser generation and for pumping. This resulted in a large amount of research on Ne atoms pumped by means of He–Ne lasers, cf., e.g., ref. [2].

The appearance of the dye laser made practically all atoms accessible for optical pumping. However, in this case a coincidence between the laser frequency and that of the pumped transition does not automatically take place. The present paper discusses the possible effects of a not precise coincidence between these frequencies.

A general theory of optical pumping of atoms by means of multimode lasers has been developed to a large extent in a series of papers by Ducloy [3–5]. In ref. [3] the author introduces the broad pumping line approximation (BLA) which makes it possible to exclude the dependence on the translation movement velocity of atoms from the equations of motion of the density matrix. So it is possible to concentrate all attention on the angular (magnetic quantum number dependent) part of absorption. It may seem that the BLA is far from realistic in the case of laser excitation. But on the other hand now an interest rises in the excitation of atoms with laser radiation with out of mode structure [6] for which the BLA holds very well. BLA assumes that independent of the velocity of the particle and of the magnitude of the applied magnetic field all the atoms, in any Zeeman state, are in sufficiently good resonance with the exciting light of large spectral width. The description method of optical pumping thus developed permitted us to perform a full analysis and interpretation of the results for a large number of experiments on Ne atoms.

A further extension of the method developed by Ducloy has recently made it possible [7–9] to show that the dynamic Stark effect may be of importance in the process of optical pumping in the BLA. We shall now present a detailed analysis of a possible manifestation of such an influence.

Using the BLA, the equations of motion for polarization moments (PM) were obtained in ref. [7], taking account of the dynamic Stark effect. In other words, the equations of motion for the expansion coefficients for the density matrix over the irreducible tensor operators [10] were obtained. They are

$$\dot{f}^K_Q = \Gamma_p \sum_{X\kappa} {}^K F^{X\kappa} \{\Phi^{(X)} \otimes \varphi^{(\kappa)}\}^K_Q + 2\mathrm{i}\omega_S \sum_{XK'} {}^K A^{XK'}_{1-} \{\Phi^{(X)} \otimes f^{(K')}\}^K_Q$$

$$- \Gamma_p \sum_{XK'} {}^K A^{XK'}_{1+} \{\Phi^{(X)} \otimes f^{(K')}\}^K_Q - (\Gamma_K - \mathrm{i}Q\omega_{J'}) f^K_Q , \tag{1a}$$

$$\dot{\varphi}^\kappa_q = -\Gamma_p \sum_{X\kappa'} {}^\kappa A^{X\kappa}_+ \{\Phi^{(X)} \otimes \varphi^{(\kappa')}\}^\kappa_q + 2\mathrm{i}\omega_S \sum_{X\kappa'} {}^\kappa A^{X\kappa'}_- \{\Phi^{(X)} \otimes \varphi^{(\kappa')}\}^\kappa_q + \Gamma_p \sum_{XK} {}^\kappa F^{XK}_1 \{\Phi^{(X)} \otimes f^{(K)}\}^\kappa_q$$

$$- (\gamma_\kappa - \mathrm{i}q\omega_{J''}) \varphi^\kappa_q + \Gamma_{j_e j_i} C_\kappa \delta_{K\kappa} \delta_{Qq} f^K_Q + \lambda^\kappa_q \delta_{\kappa 0} \delta_{q0} . \tag{1b}$$

Here f^K_Q and φ^κ_q denote PM of rank K and κ, respectively, describing the excited j_e and ground j_i state of the atom, Γ_K and γ_κ are their relaxation rates, $\omega_{J'}$ as well as $\omega_{J''}$ are the frequencies of the Zeeman splitting of states j_e and j_i in an external magnetic field. The first summand in the right-hand part of both equations describes the absorption of light. The absorption rate equals [11]

$$\Gamma_p = 2\pi (2j_e + 1)^{-1} \hbar^{-2} |(j_e \| r \| j_i)|^2 e^2 i(\omega_0) , \tag{2}$$

where $(j_e \| r \| j_i)$ is the reduced matrix element, e is the electron charge, $i(\omega_0)$ is the spectral density of the laser radiation at the atomic transition frequency ω_0. The coefficients ${}^K F^{X\kappa}$, ${}^\kappa F^{XK}_1$, ${}^\kappa A^{X\kappa'}_\pm$, ${}^K A^{XK'}_{1\pm}$, which account for the conservation of the angular momentum in the absorption and emission of a photon, can be found from

$$ {}^K F^{X\kappa} = \frac{(2j_e+1)^{3/2}(2X+1)(2\kappa+1)}{(2j_i+1)^{1/2}(2K+1)^{1/2}} (-1)^{X+1} \begin{Bmatrix} K & j_e & j_e \\ X & 1 & 1 \\ \kappa & j_i & j_i \end{Bmatrix} , \tag{3}$$

$$ {}^\kappa F^{XK}_1 = \frac{(2j_e+1)^{1/2}(2j_i+1)^{1/2}(2X+1)(2K+1)}{(2\kappa+1)^{1/2}} (-1)^{X+1} \begin{Bmatrix} \kappa & j_i & j_i \\ X & 1 & 1 \\ K & j_e & j_e \end{Bmatrix} , \tag{4}$$

$$ {}^\kappa A^{X\kappa'}_\pm = \frac{1 \pm (-1)^{\kappa+X+\kappa'}}{2} \frac{(2j_i+1)(2X+1)(2\kappa'+1)}{(2\kappa+1)^{1/2}} (-1)^{j_e - j_i + \kappa'} \begin{Bmatrix} \kappa & X & \kappa' \\ j_i & j_i & j_i \end{Bmatrix} \begin{Bmatrix} 1 & 1 & X \\ j_i & j_i & j_e \end{Bmatrix} , \tag{5}$$

$$ {}^K A^{XK'}_{1\pm} = \frac{1 \pm (-1)^{K+X+K'}}{2} \frac{(2j_e+1)(2X+1)(2K'+1)}{(2K+1)^{1/2}} (-1)^{j_e - j_i + K'} \begin{Bmatrix} K & X & K' \\ j_e & j_e & j_e \end{Bmatrix} \begin{Bmatrix} 1 & 1 & X \\ j_e & j_e & j_i \end{Bmatrix} , \tag{6}$$

where the quantities inside the accolades are $9j$ and $6j$ symbols [12]. The polarization of the exciting light is denoted by the Dyakonov [13] tensor

$$\Phi^X_\xi = (2X+1)^{-1/2} \sum_{q_1 q_2} (-1)^{q_2} E^{q_1} (E^{q_2})^* C^{X\xi}_{1-q_1 1 q_2} , \tag{7}$$

where $C^{c\gamma}_{a\alpha b\beta}$ are the Clebsch–Gordan coefficients, e_{q_1}, e_{q_2} are cyclic components of the unit vector of light polarization, and $\otimes$ denotes irreducible tensor multiplication.

The second summands in eqs. (1) are responsible for the dynamic Stark effect which causes a shift in the resonance frequency of the atomic transition by [11]

$$\omega_S = \frac{|(j_e \| r \| j_i)|^2}{\hbar^2 (2j_e+1)} e^2 \int \frac{i(\omega_l)}{\omega_l - \omega_0} \, \mathrm{d}\omega_l . \tag{8}$$

It may be seen from (8) that ω_S differs from zero in the case when the resonance absorption frequency is situated out of center in the spectral contour of the laser $i(\omega_l)$.

The third summands describe stimulated emission of light. This process is the reverse of absorption.

The fourth summands describe the relaxation of the PM, including their destruction by the external magnetic field.

The fifth summand of eq. (1b) characterizes the reverse spontaneous transitions $j_e \to j_i$ at a rate $\Gamma_{j_e j_i}$. In the general case $\Gamma_{j_e j_i}$ does not coincide with Γ_K, since the latter includes also collisional relaxation and possible transitions on other levels, different from j_i. Conservation of angular momentum in reverse spontaneous transitions is provided by the coefficient

$$C_\kappa = (-1)^{j_e+j_i+\kappa+1}(2j_e+1)^{1/2}(2j_i+1)^{1/2}\begin{Bmatrix} j_i & j_i & \kappa \\ j_e & j_e & 1 \end{Bmatrix}. \tag{9}$$

Finally, the last summand of eq. (1b) describes the restoration of the lower level j_i population in isotropic relaxation processes.

The main difference of the equations of motion of the PM from those used previously, cf., e.g., ref. [4], consists in the appearance of terms proportional to ω_S, which are responsible for the action of the dynamic Stark effect. We shall now try to demonstrate what new effects this phenomenon may lead to.

In the absence of the dynamic Stark effect only alignment of atoms, both in excited and ground states, is known to take place on excitation with linearly polarized light. In other words, the atomic ensemble is characterized by PM of even ranks K, κ, and no orientation or odd rank PM arise, cf., e.g., ref. [2]. This is formally due to the fact that Φ_ξ^X differs from zero only at $X=0$, 2 for linearly polarized light, whilst the coefficients ${}^K F^{X\kappa}$, ${}^\kappa F_1^{XK}$, ${}^\kappa A_+^{X\kappa'}$ and ${}^K A_{1+}^{XK'}$ responsible for PM formation differ from zero only in the case of an even sum of the upper indices. The situation changes under the conditions of the dynamic Stark effect ($\omega_S \neq 0$), and the coefficients ${}^\kappa A_-^{X\kappa'}$ and ${}^K A_{1-}^{XK'}$ which differ from zero at an odd sum of the upper indices start playing a role. Transition from alignment to orientation starts, which manifests itself experimentally in the form of the appearance of circularly polarized fluorescence, in the transition $j_e \to j_i$.

Let us discuss in more detail the following model situation. Let us assume that the exciting radiation is sufficiently weak, so as not to induce the stimulated transitions $\Gamma_p \ll \Gamma_K$, however, strong enough for the optical pumping of atoms $\Gamma_p \leqslant \gamma_\kappa$. The magnitude of the orientation on the lower level j_i can then be estimated expanding the solution for the PM into a series over the parameters Γ_p/γ_κ and ω_S/γ_κ. If we put the angle between the polarization vector of the exciting light $\boldsymbol{E}$ and the external magnetic field equal to θ, then we have for the transitions $j_i = 1$–$j_e = 1$ in second order of the expansion,

$$^{(2)}\varphi^1_0 = \frac{\sqrt{2}}{12}\frac{\omega_S\Gamma_p}{\gamma_1}\sin^2\theta\left(\frac{\omega'\sin^2\theta}{\gamma_2^2+4\omega_{j_i}^2} + \frac{\omega''\cos^2\theta}{\gamma_2^2+\omega_{j_i}^2}\right), \tag{10}$$

where

$$\omega' = \omega_{j_i} + \frac{1}{2}\frac{\Gamma_{j_e j_i}(\omega_{j_e}\gamma_2 + \omega_{j_i}\Gamma_2)}{\Gamma_2^2+4\omega_{j_e}^2}, \qquad \omega'' = \omega_{j_i} + \frac{1}{2}\frac{\Gamma_{j_e j_i}(\omega_{j_e}\gamma_2+\omega_{j_i}\Gamma_2)}{\Gamma_2^2+\omega_{j_e}^2},$$

and

$$^{(2)}\varphi^1_{\pm1} = \pm \mathrm{i}\,\mathrm{e}^{\pm\mathrm{i}\varphi}\frac{\omega_S\Gamma_p\sin^2\theta}{48(\gamma_1 \mp \mathrm{i}\omega_{j_e})}\left[\sin^2\theta\left(\frac{o_1}{\gamma_2 \mp 2\mathrm{i}\omega_{j_i}} - \frac{o_2^*}{\gamma_2 \pm \mathrm{i}\omega_{j_i}}\right) + (3\cos^2\theta - 1)\left(\frac{o_2}{\gamma_2 \mp \mathrm{i}\omega_{j_i}} - \frac{o_3}{\gamma_2}\right)\right],$$

$$o_1 = 1 + \frac{1}{2}\frac{\Gamma_{j_e j_i}}{\Gamma_2 \mp 2\mathrm{i}\omega_{j_e}}, \qquad o_2 = 1 + \frac{1}{2}\frac{\Gamma_{j_e j_i}}{\Gamma_2 \mp \mathrm{i}\omega_{j_e}}, \qquad o_3 = 1 + \frac{1}{2}\frac{\Gamma_{j_e j_i}}{\Gamma_2}. \tag{11}$$

It may be seen that in the case of orthogonality between the vector $\boldsymbol{E}$ and the magnetic field, i.e. $\theta = \frac{1}{2}\pi$, only the longitudinal orientation φ_0^1 is formed, i.e. orientation along the magnetic field. In the opposite case, transversal orientation $\varphi^1_{\pm1}$ is also present in the ensemble. It ought to be stressed that a transition from alignment to orientation takes place only in the presence of the magnetic field ω_{j_i}, $\omega_{j_e} \neq 0$, and if the dependence of the effect on ω_{j_i} and ω_{j_e} is of resonance character.

If we have arbitrary angular momentum values j_i, j_e, and at $\theta=\frac{1}{2}\pi$ the longitudinal orientation appearing in second order of the expansion is connected with the transversal alignment ${}^{(1)}\phi^{2}_{-2}$ calculated in first order,

$$ {}^{(2)}\phi^{1}_{0}=-\frac{\omega_S}{\gamma_1}\,{}^{1}A^{22}_{-}C^{10}_{222\,-2}\times 2\,\mathrm{Im}(\Phi^{2}_{2}\,{}^{(1)}\phi^{2}_{-2})\;. \qquad (12)$$

Since the coefficient ${}^{1}A^{22}_{-}$ is proportional to j_i^{-1} in the limit of high j_i values, expression (12) permits one to conclude that the transition effect from alignment to orientation is purely of quantum nature and disappears at the transition in the limit $j_i\to\infty$.

In the case of the transition $j_i=1$–$j_e=2$ the formula yields

$$ {}^{(2)}\phi^{1}_{0}=\frac{\sqrt{2}}{108}\frac{\omega_S\Gamma_p}{\gamma_1(\gamma_2^2+4\omega_{j_i}^2)}\left(\omega_{j_i}-\frac{7\Gamma_{j_ej_i}(\omega_{j_e}\gamma_2+\omega_{j_i}\Gamma_2)}{2(\Gamma_2^2+4\omega_{j_e}^2)}\right). \qquad (13)$$

Comparison between expressions (10) at $\theta=\frac{1}{2}\pi$ and (13) shows that reverse spontaneous transitions at the rate $\Gamma_{j_ej_i}$ may enhance ($j_i=1$–$j_e=1$) or weaken, and even (at $\Gamma_{j_ej_i}\approx\Gamma_2$) change the sign of the effect of transition from alignment to orientation. In other words, the second summand between the large parentheses in expression (13) may turn out to be larger than the first one.

Experimental observation of the orientation thus formed is connected with the circular polarization of fluorescence along the magnetic field. The intensity of the fluorescence I with a polarization $\boldsymbol{E}'$ in the transition j_1–j_0 is expressed by [13]

$$ I(E')\sim(-1)^{j_1+j_0}(2j_1+1)^{1/2}\sum_{K=0}^{2}(2K+1)\begin{Bmatrix}1 & 1 & K\\ j_1 & j_1 & j_0\end{Bmatrix}\sum_{Q=-K}^{K}(-1)^{Q}\rho^{K}_{Q}\Phi^{K}_{-Q}(\hat{E}')\;, \qquad (14)$$

where ρ^{K}_{Q} is the PM on level j_1. If the observation is carried out along the magnetic field, then the degree of circular polarization equals

$$ C=\frac{I_r-I_\ell}{I_r+I_\ell}\sim\rho^{1}_{0}\;, \qquad (15)$$

where I_r, I_ℓ are the right and left polarized components, respectively. In these examples it is possible to observe fluorescence from j_i to some lower situated level j_f. Then we have $\rho^1_0=\varphi^1_0$ in expression (15). If j_i represents the ground state of the atom, and fluorescence from it cannot be directly observed, the effect can nevertheless manifest itself by the following scheme. Orientation produced by the exciting light on level j_i according to eq. (1a) is transferred to the excited level j_e on which the PM $f^{1}_{0}\sim\varphi^1_0$ is produced. The observation is carried out on the transition $j_e\to j_f$ and we have $\rho^1_0=f^1_0$ in formula (15).

The above analysis makes it possible to obtain a clear idea of the mechanism of the transition from alignment to orientation of atoms in a magnetic field under the action of the dynamic Stark effect. The physical reason for the appearance of the orientation is the following. On the excitation of atoms by intensive plane polarized light in a weak magnetic field the coherence of the magnetic sublevels of the states j_i, j_e appears [2]. An increase in the magnetic field intensity leads to a spread in the Zeeman components and to the destruction of the coherence. This spread proceeds with equal efficiency for all magnetic sublevels in the absence of the dynamic Stark effect, independently of the value of the magnetic quantum number, owing to the equidistance of all the Zeeman components. If, however, the dynamic Stark effect occurs, the equidistance of the magnetic sublevels does not exist any longer [14,15], and the coherence for the positive and negative magnetic sublevels disappears with a different efficiency. This is the reason which leads to the conversion of alignment into orientation. The modification of the Zeeman structure due to the dynamic Stark effect was experimentally studied and theoretically analyzed using an effective Hamiltonian formalism by Cohen-Tannoudji and Dupont-Roc [14]. Such a transformation of alignment into orientation in a discharge experiment with argon ions was experimentally studied in a recent paper of Elbel et al. [16]. In the discharge conditions there are problems to separate the influence of the dynamic Stark effect and the collisional interaction with electrons. The authors suggest that in their experiments the influence of electrons prevails.

For a more exact, but less comprehensible analysis of the transformation of alignment into orientation as a result of the dynamic Stark effect, it is necessary to solve the system of equations (1) numerically.

As an example we shall analyze the optical pumping of the transition $2p_4$–$3s_2$ ($j_i=2$–$j_e=1$) for a Ne atom. The authors in ref. [2] observed optical pumping on this transition by means of excitation with a He–Ne laser. The geometry was chosen such that the vector $\boldsymbol{E}$ of laser radiation and the magnetic field $\boldsymbol{H}$ were oriented orthogonally, cf. fig. 1, whilst the signal was observed as the intensity difference $I_{\parallel}-I_{\perp}$ between two fluorescence components in the transition $3s_2$–$2p_1$ ($j_e=1$–$j_f=0$). Since the pumped atoms and the atoms from which the laser generation takes place are of the same sort in this case, we have $\omega_S=0$. In the $\boldsymbol{H}$ dependence of $I_{\parallel}-I_{\perp}$ as observed in the experiments, a peak appeared in the vicinity of the zero magnetic field, cf. curve 1 in fig. 2. This peak was interpreted as a manifestation of the hexadecapole moment φ_q^4 of the lower level j_i. Assume the following values of the dynamic constants, $\gamma_0=9.8\ \mu s^{-1}$, $\gamma_2=\gamma_4=14.5\ \mu s^{-1}$, $\Gamma_0=2.75\ \mu s^{-1}$, $\Gamma_2=5.75\ \mu s^{-1}$, $\Gamma_{j_e j_i}=0.5\ \mu s^{-1}$, $\Gamma_p=115\ \mu s^{-1}$, $\omega_{j_i}=1.299\times\mu_B H/\hbar$, $\omega_{j_e}=1.293\times\mu_B H/\hbar$, where μ_B is the Bohr magneton. An excellent coincidence of the theoretical curve with the experimental results was obtained [2] by solving a system of equations similar to (1) without accounting for the dynamic Stark effect. For the calculation of the observed signal formula (14) was employed. The results of the calculations repeated by us are presented by curve 1 of fig. 2.

Let us now assume that a similar experiment is performed by means of a dye laser, and the dynamic Stark effect is present due to the inexact tuning of the laser frequency on the atomic transition frequency, such that $\omega_S=\Gamma_p$. Leaving all parameters unchanged in the calculation but adding $\omega_S=115\ \mu s^{-1}$ we obtain curve 2 of fig. 2. It may be seen that the dynamic Stark effect may substantially distort the signal if the observation is carried out with respect to the linearly polarized light. It ought to be noted that the situation, when ω_S equals Γ_p in magnitude or even exceeds it, is fully realistic, as may be seen from a comparison of formulas (2) and (8). This problem is discussed in ref. [9] in more detail.

In order to observe directly the orientation obtained on the levels j_i and j_e we may calculate the signal I_r-I_ℓ as viewed along the direction of the magnetic field, cf. fig. 1. Repeating the calculations for the same parameter

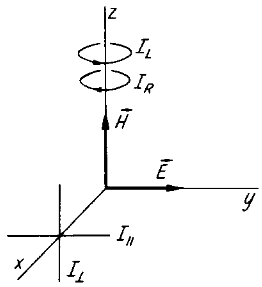

Fig. 1. Geometry for the calculations.

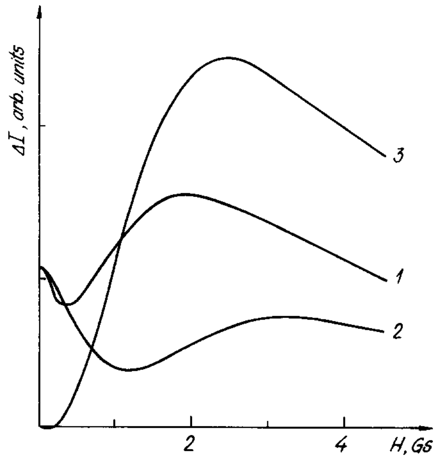

Fig. 2. Dependence of the difference between the two polarization components in the fluorescence on the external magnetic field H. (1) Absence of the dynamic Stark effect, linearly polarized observation; (2) presence of the dynamic Stark effect, linearly polarized observation; (3) presence of the dynamic Stark effect, circularly polarized observation.

values as for curve 2, we obtain curve 3 of fig. 2. In this case the amplitude of the expected signal exceeds that of the signal observed in the case of linearly polarized light.

Thus, accounting for the action of the dynamic Stark effect on optical pumping of atoms may be essential for a correct interpretation of the experimental results. Such an action may be observed in the transition from alignment to orientation. This, as shown above, is a nonlinear effect which disappears at states with high angular momentum values.

Owing to its large amplitude, the registration of signals of the type as in curve 3 of fig. 2 might be of independent interest in the studies of atoms and in the determination of various atomic constants on which these signals are dependent.

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