OPTICAL BACK–GOUDSMIT EFFECT: LASER DECOUPLING OF HYPERFINE INTERACTIONS IN ATOMS*

Wojciech Gawlik and Jerzy Zachorowski

M. Smoluchowski Institute of Physics, Jagellonian University
Reymonta 4, 30-059 Kraków, Poland

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Laser perturbation of an atom with fine or hyperfine structure is analyzed. The use of sufficiently powerful, or spectrally broad light produces effects, which form an optical analogue to the Back–Goudsmit effect. Such laser decoupling of hyperfine interaction is easily understood in terms of an analogy of the level-crossing effect and the double-slit experiment. The consequences of the optical Back–Goudsmit effect for efficiency of optical pumping in ^{3}He are discussed.

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1. Introduction

Interesting effects occur in a physical system when two, or more, perturbations of a comparable strengths act upon it. A familiar example in atomic physics is when interaction of an external magnetic field with electronic spins is about as strong as the intra-atomic spin-orbit interaction responsible for the atomic fine structure (fs) or the interaction of a nucleus with an electronic shell, which produces the hyperfine structure (hfs). These effects are known as the Paschen–Back [1] and Back–Goudsmit [2] effects, respectively.

The paper revisits previous observations of the effect of laser decoupling of hyperfine interaction on atomic-state interference [3–5] and then discusses possible consequences of optical pumping with strong light on nuclear polarization of ^{3}He, used for the magnetic resonance imaging [6].

As lasers are widely used in atomic physics experiments nowadays, it is appropriate to check how strongly they perturb atomic energy-level structure and whether they can act similarly to the magnetic field in the above mentioned classical examples. Fig. 1 demonstrates that the available lasers

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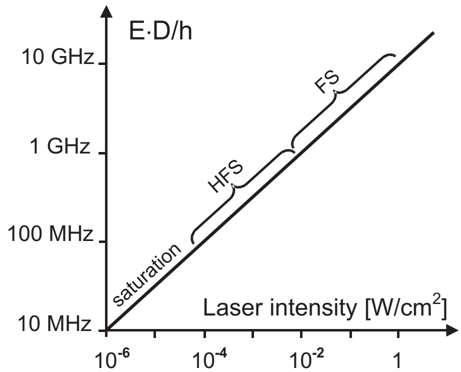

Fig. 1. Rabi frequency of atom-light interaction *versus* laser intensity for a typical allowed optical transition compared to other intra-atomic interactions.

are capable of producing perturbations sufficient not only to saturate the atomic transition, but also comparable with the hyperfine or fine structure, such that $\boldsymbol{E}\cdot\boldsymbol{D}\approx\boldsymbol{J}\cdot\boldsymbol{I}$ or $\boldsymbol{E}\cdot\boldsymbol{D}\approx\boldsymbol{L}\cdot\boldsymbol{S}$, where $\boldsymbol{E}$ is the electric field of the light wave and $\boldsymbol{D}$ is the atomic dipole moment, $\boldsymbol{L}$ and $\boldsymbol{J}$ are orbital and full electronic angular momenta, respectively. $\boldsymbol{S}$ and $\boldsymbol{I}$ represent electronic and nuclear spin, respectively. The strength of the light-atom interaction is usually characterized by the value of the Rabi frequency $\Omega=\boldsymbol{E}\cdot\boldsymbol{D}/h$.

The analysis of the light perturbation of atomic energy level structure associated with the fine and/or hyperfine interaction can be performed in a standard way, *i.e.*, by diagonalizing the full atomic Hamiltonian. Such treatment has been applied in many cases, *e.g.*, [7]. Here we wish to present a simple, intuitive model that allows understanding of experimental results in terms of atomic state interference.

2. Atomic state interference

We would like to discuss several consequences of such strong perturbation by laser light starting with some experiments on atomic state interference. To get some insight into the atomic-state interference we first discuss a three-level atomic system consisting of the ground state g with angular momentum $J=0$ and the excited state with $J'=1$. Resonant light of a linear σ polarization causes excitation only of the excited-state Zeeman sublevels $m'=\pm1$. The $m'=0$ upper-state sublevel is not excited and can be ignored in the analysis, so the system $\{g,\, m'=-1,\, m'=+1\}$ behaves like a three-level, V-like atomic structure (Fig. 2(a)). Levels $m'=\pm1$ can decay back to the g state or any other final state g', as depicted by broken lines in Fig. 2(a).

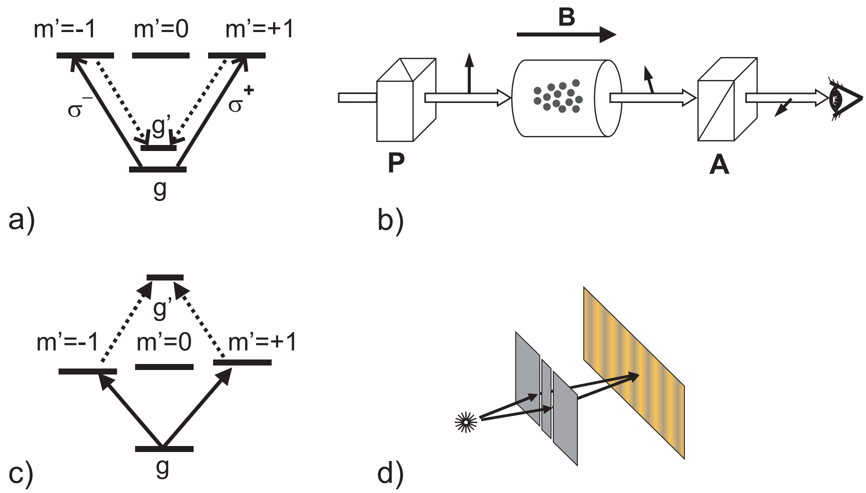

Fig. 2. (a) Atomic structure consisting of a ground levels g and g' with $J = 0$ and two excited sublevels with $J' = 1$, $m' = \pm 1$ interacting with linear σ-polarized light; (b) scheme of the Faraday effect experiment: P and A are polarizers, B indicates the magnetic field direction; (c) light scattering can be compared to (d) classical double-slit Young's experiment when the final ground state is flipped above the excited state. Different trajectories between g and g' via the $m' = \pm 1$ sublevels correspond to transitions through two slits.

2.1. Observations of the atomic state interference in the resonant Faraday effect.

Atomic state interference can be observed in various ways. First experiment of this kind has been performed by Hanle [8] who studied depolarization of the resonant fluorescence. Another possibility is the Faraday effect observed in the geometry of Fig. 2(b). Results of such experiments can be interpreted in terms of the classical interference experiment. Scattering of linearly polarized photons on atomic system such as in Fig. 2(a) proceeds along two quantum pathways between initial and final states g and g' via two intermediate states $m' = \pm 1$ (Fig. 2(c)). This corresponds to two optical trajectories in the double-slit Young's experiment (Fig. 2(d)). The phase difference responsible for the interference pattern is controlled by the magnetic-field splitting of the two relevant m' sublevels in the case of the atomic-state interference and this determines the Faraday rotation angle. In particular, the interference is the strongest when the m' sublevels are degenerate around the magnetic field $B = 0$ and disappears as the level degeneracy is lifted, since then the m' sublevels are no more degenerate and the quantum trajectories lose their indistinguishability.

The first experiment of the nonlinear Faraday effect with a narrowband laser light has been performed by Gawlik *et al.* [9] on sodium vapor. The interference of quantum trajectories between Zeeman sublevels yields a narrow feature in the scattering signal around $B = 0$. This feature is seen in experimental recordings for different light intensities and for both resonant lines of sodium in Fig. 3(a). Fig. 3(b) depicts results of the similar exper-

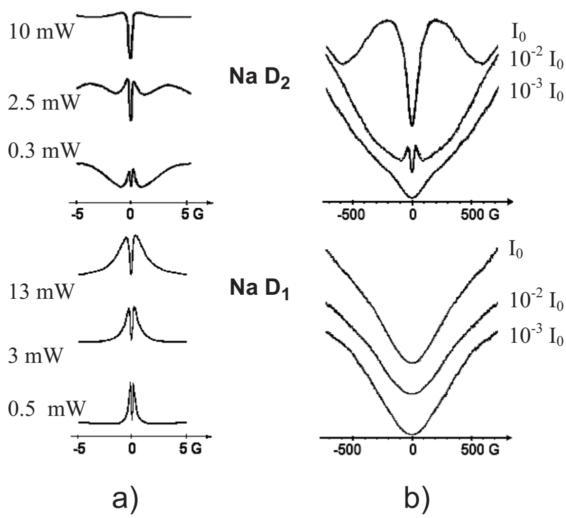

Fig. 3. Faraday effect signal: transmitted light intensity *versus* magnetic field for different light intensities and for both resonant sodium lines: (a) monochromatic cw laser [9], (b) broadband, pulsed laser [4]. The signal shows narrow features around $B = 0$ due to atomic-state interference and a broad background of the linear Faraday effect. For monochromatic laser the narrow features occur for both lines, for powerful, broadband excitation thy appear for D_2, but not for the D_1 line.

iment performed with a broadband laser by Gawlik and Zachorowski [4]. One can observe the narrow interference features around $B = 0$ for both Na resonance lines D_1 ($3^2S_{1/2}$–$3^2P_{1/2}$) and D_2 ($3^2S_{1/2}$–$3^2P_{3/2}$) in the case of the narrowband laser, but only for the D_2 line, and not for the D_1 line with the broadband laser.

2.1.1. Interpretation

As the hyperfine splittings of the sodium $^2P_{1/2}$ and $^2P_{3/2}$ states are of the same order (Fig. 4), the absence of the interference pattern for a strong broadband excitation of the Na D_1 line cannot be related to the size of the hyperfine structure.

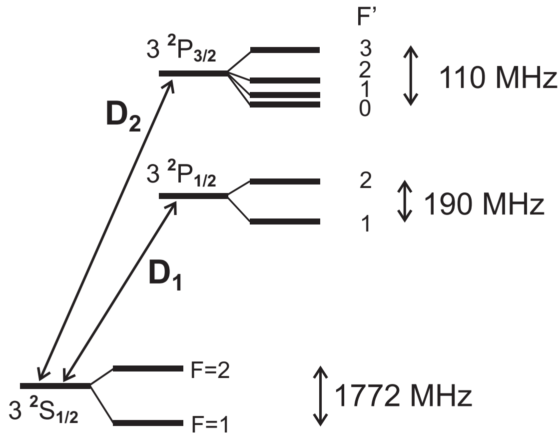

Fig. 4. Energy-level structure of sodium D lines.

The simplest possible atomic structure, where explanation of the results shown in Fig. 3 is possible, is the model of an atom with $I = 1/2$ and $J = 1/2$ or $3/2$ [3, 5]. Interpretation taking into account the exact energy-level structure of sodium has been presented in [4]. In our simple model, the D_1 line excited by a linearly σ-polarized light consists of transitions shown in Fig. 5(a) in the $|J, I, F, m_F\rangle$ representation (labelled $|F, m_F\rangle$ for compactness). If one ignores the hfs coupling, then the transitions can also be shown in the $|J, m_J, I, m_I\rangle$ basis as in Fig. 5(b) (in short the $|m_J, m_I\rangle$ basis).

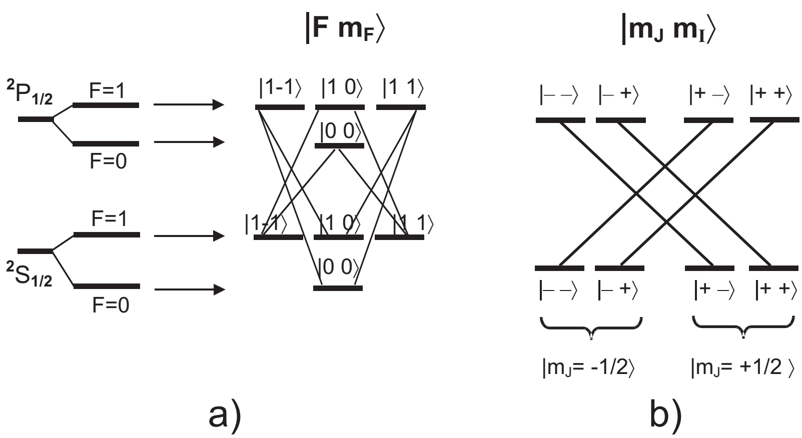

Fig. 5. Energy-level structure of an atom with $J = J' = 1/2$ and $I = 1/2$ (a) in the $|F, m_F\rangle$ representation, (b) in the $|m_J, m_I\rangle$ basis.

If the $\boldsymbol{J}\cdot\boldsymbol{I}$ hyperfine coupling is taken into account, the states with different m_J, m_I values are coupled as shown by broken lines in Fig. 6(a). When spontaneous emission can be neglected (case of a strong light intensity), the structure of Fig. 6(a) splits into two independent four-level substructures (Fig. 6(b)) and each of them can be analyzed separately.

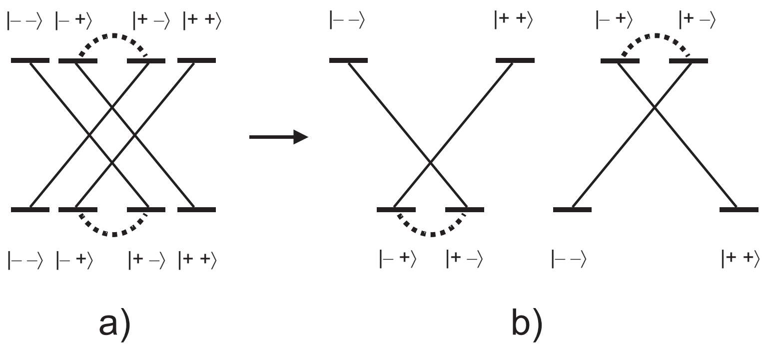

Fig. 6. (a) The energy-level structure of Fig. 5(b) with hyperfine coupling indicated by broken lines. (b) When spontaneous emission can be neglected the structure consists of two separate subsystems.

As an example, we take a closer look into light scattering process in one of these structures Fig. 7(a). Similarly to the three-level system discussed above (Fig. 2), the process can be analyzed in terms of an interference experiment Fig. 7(b).

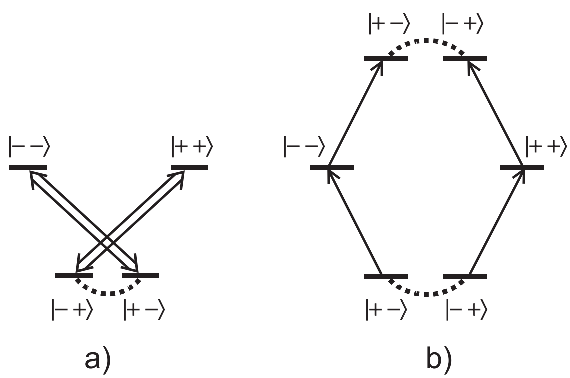

Fig. 7. (a) Single substructure of Fig. 6(b) and (b) its equivalent form corresponding to the double-slit interference.

There are two distinct limits of the competition between the light $\boldsymbol{E}\cdot\boldsymbol{D}$ and hyperfine $\boldsymbol{J}\cdot\boldsymbol{I}$ interactions. If $\boldsymbol{E}\cdot\boldsymbol{D} \ll \boldsymbol{J}\cdot\boldsymbol{I}$, the states coupled by the strong hyperfine interaction are degenerate and well characterized by quantum numbers F, m_F. This corresponds to the situation when hfs is well spectrally resolved. Optical transitions induced by the linearly polarized light are then between well-defined initial and final states with $m_F = 0$ via two intermediate states $m_F = \pm 1$ (Fig. 8(a)). This is analogous to the Young's experiment when it cannot be specified through which slit the photon has been transmitted (Fig. 8(b)) and results in interference pattern around $B = 0$ such as in Fig. 3(a).

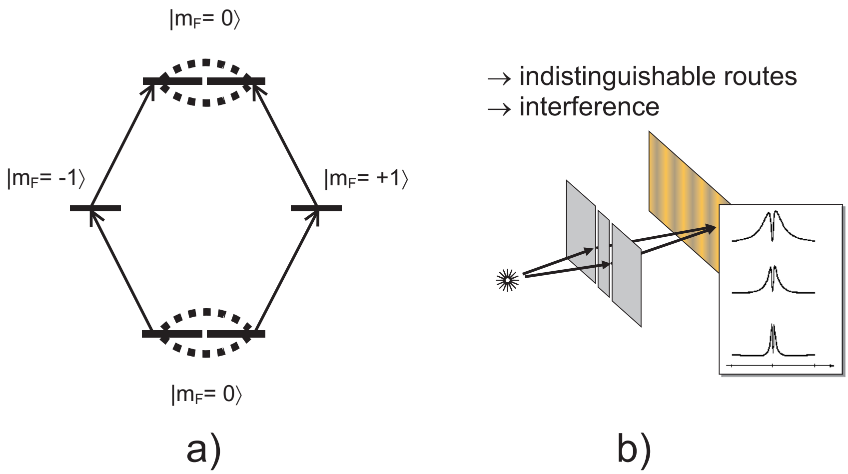

Fig. 8. (a) Atomic interference for strong hyperfine coupling: F, m_F are good quantum numbers, (b) interference occurs and shows up as narrow features in Faraday effect signal (inset).

In the opposite case, *i.e.*, when $\boldsymbol{E} \cdot \boldsymbol{D} \gg \boldsymbol{J} \cdot \boldsymbol{I}$, the hyperfine coupling can be neglected. This corresponds to the situation when power broadening (on the order of the Rabi frequency Ω) makes the hyperfine structure unresolved. The light-induced transitions proceed on two independent pathways (Fig. 9(a)). The initial and final states for both pathways are no longer degenerate, which corresponds to the case when the photon trajectories in the Young's experiment can be identified resulting in no interference pattern. In the scattering experiment this corresponds to signals without narrow features at $B = 0$, such as seen in lower part of Fig. 3(b) shown as an inset in Fig. 9(b).

The analogy with the interference explains also the fact that for the Na D_2 line there is interference feature around $B = 0$, despite strong intensity and wide spectral width of the laser (upper part of Fig. 3(b)). As seen in Fig. 10(a) there are two sets of transition pathways for the D_2 line that can interfere within each set: the first one with initial and final states $m_J = -1/2$ and two indistinguishable intermediate states $m_J = -3/2$ and $+1/2$,

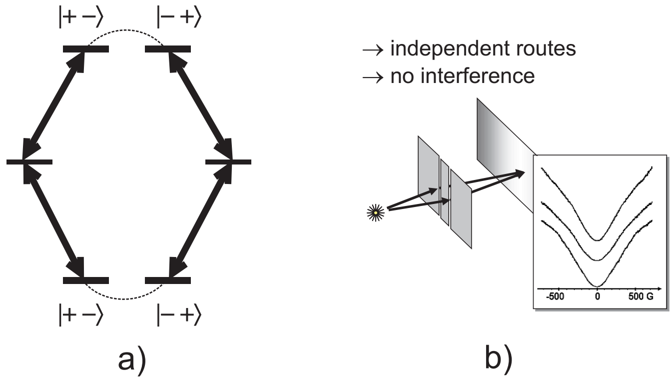

Fig. 9. (a) Atomic interference for weak hyperfine coupling: m_J, m_I are good quantum numbers, (b) interference does not show up: narrow features are absent in Faraday effect signal (inset).

the second one having initial and final states with $m_J = +1/2$ and two indistinguishable intermediate states $m_J = -1/2$ and $+3/2$. In that case, the interference pattern is well visible (Fig. 10(b)).

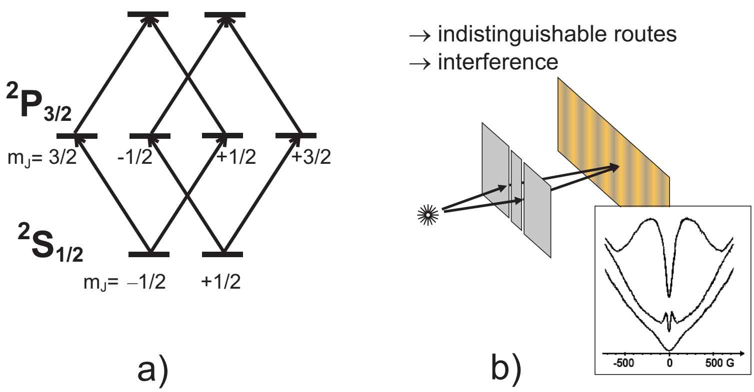

Fig. 10. (a) Atomic structure for sodium D_2 line with powerful, broadband laser. Even in the $|m_J, m_I\rangle$ basis there are indistinguishable pathways, (b) interference occurs and shows up as narrow features in Faraday effect signal (inset).

3. Effect of the optical Back–Goudsmit effect on nuclear spin polarization of ^{3}He

Having discussed the effects of the optical Back–Goudsmit effect on the atomic state superpositions or interference, we turn now to the analysis whether such an effect could be important in redistribution of atomic state populations, *i.e.*, in optical pumping. As pointed out by Nacher [6] in this issue, high degree of nuclear spin polarization of ^{3}He can be obtained by optical pumping with laser tuned to the $\lambda = 1083$ nm transition between the metastable state 2^3S_1 and the $2^3P_{0,\,1,\,2}$ states (Fig. 11(a)) taking advantage of the metastability exchange collisions [10]. Since the ^{3}He nucleus has spin $I = 1/2$, the states have hyperfine structure with many sublevels characterized by various F quantum numbers. In ^{3}He, the magnitude of the hyperfine structure is comparable with the fine structure, which additionally complicates the discussion of the light perturbation. For a qualitative analysis, however, we concentrate here exclusively on the hyperfine structure decoupling. Fig. 11(b) depicts the fine and hyperfine structure of the

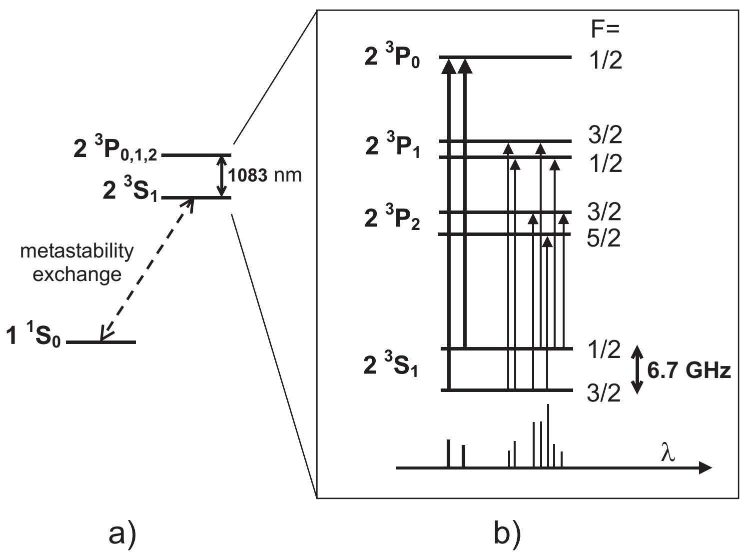

Fig. 11. (a) Structure of energy levels of ^{3}He involved in optical pumping leading to nuclear spin polarization. (b) Transitions between various fine and hyperfine structure states contributing to the 1083 nm line.

1083 nm line. Optical pumping of the metastable state is performed essentially by two components that excite the 2^3P_0 state. Fig. 12 shows the optical pumping transitions induced by the circularly σ^+ polarized light in more detail. All relevant sublevels are labeled with the m_F quantum numbers. They are also characterized by the appropriate m_J and m_I quantum numbers ($m_J = -1,\ 0,\ 1$ and $m_I = \pm 1/2$ which is symbolized by an arrow pointing up or down). In the $|F,\, m_F\rangle$ representation it may be easily shown that the $m_F = +3/2$ sublevel is the one the most efficiently pumped by the σ^+ light. If hyperfine coupling is strong enough that F and m_F are good quantum numbers, the $m_F = +3/2$ sublevel is not coupled to any other state by the $\boldsymbol{J} \cdot \boldsymbol{I}$ interaction and remains a pure state. Since this sublevel

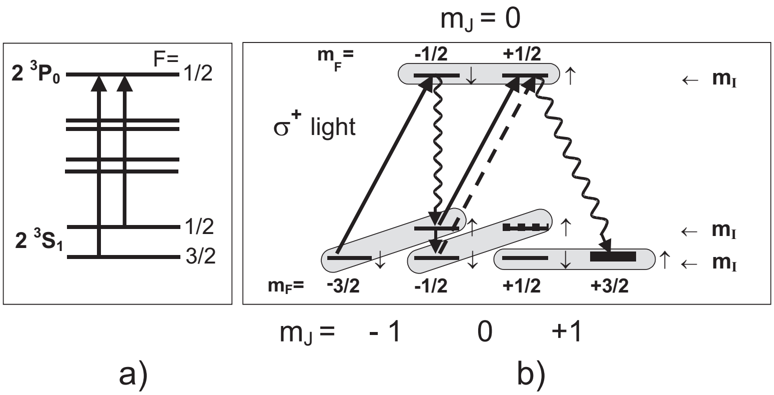

Fig. 12. Transitions most efficient in creation of nuclear polarization by optical pumping of ^{3}He with the 1083 line. (b)— Optical pumping by the σ^+ light exciting the 2^3P_0 state as described in text. Straight arrows are transitions induced by the pumping light (broken line mark a weaker component). Wavy arrows represent spontaneous emission channels relevant for populating the $m_F = +3/2$ sublevel. The grey ovals link the m_F sublevels that can be associated with separate m_J values. The $\pm 1/2$ values of m_I are indicated by small arrows.

corresponds to $m_I = +1/2$, strong optical pumping of this sublevel is equivalent to strong polarization of the nuclear spin $\boldsymbol{I}$. In Fig. 12(b) the pairs of sublevels that contribute to given values of m_J are also marked by shaded oval contours. If the optical perturbation $\boldsymbol{E} \cdot \boldsymbol{D}$ is sufficiently strong, F and m_F cease to be good quantum numbers and the uncoupled basis $|m_J, m_I\rangle$ becomes more appropriate. Fig. 13 depicts the same sublevels involved in the optical pumping, as in Fig. 12(b), but now rearranged in pairs that con-

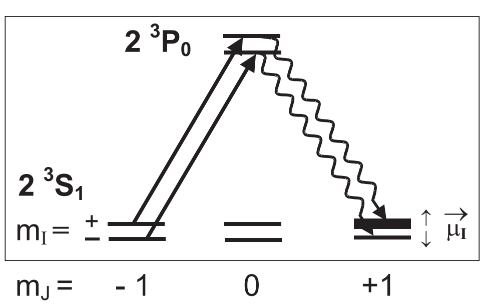

Fig. 13. Optical pumping by the σ^+ light of high intensity exciting the 2^3P_0 state. The states are shown in the $|m_J, m_I\rangle$ basis. Optical pumping populates most efficiently state $m_J = +1$, which is not a pure state of the nuclear spin. Thus, nuclear spin polarization is reduced.

tribute to given m_J values (marked by ovals in Fig. 12(b)). It can be seen that in the $|m_J, m_I\rangle$ basis, the state that is the most efficiently pumped is the $m_J = +1$ state, which is no longer a pure state, but acquires admixture of $m_I = -1/2$. This means that the degree of nuclear polarization is reduced when hyperfine structure is uncoupled.

4. Conclusions

In conclusion, we would like to point out that the optical analogue to the Back–Goudsmit (Paschen–Back) effect is quite feasible with the available laser powers. One can achieve the situation when the strength of the $\boldsymbol{E} \cdot \boldsymbol{D}$ interaction is bigger that the intra-atomic interaction $\boldsymbol{I} \cdot \boldsymbol{J}$ ($\boldsymbol{L} \cdot \boldsymbol{S}$). In such case, the power broadening of the optical transition reduces the resolution to the extent that the hyperfine (fine) structure of the perturbed spectral line becomes unresolved. Similar consequences occur also in the case that has not been explicitly discussed above, when the laser light is not monochromatic and its width is bigger than the hyperfine (fine) structure. Such case is reported in [4], where no interference signal has been detected, although it was well visible with monochromatic laser, when the hfs structure has been resolved. The effect cannot be attributed to the power broadening as it remained even after reduction of the power of the broadband laser. The two factors may act jointly, *e.g.*, in case of pulsed and powerful lasers, making the whole interaction more complicated. Additional complications arise when the fine and hyperfine structures are comparable, as it is the case of the 1083 nm transition in ^{3}He.

The occurrence of the optical Back–Goudsmit or Paschen–Back effects can significantly alter the result of the light-atom interaction. The examples discussed above are cancellation of the atomic-state interference or reduction of the optical pumping efficiency.

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