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On the experimental measurement of the photon's fundamental static magnetic field operator $\hat{B}_{\Pi}$: the optical Zeeman effect in atoms

M.W. Evans

732 Theory Center, Cornell University, Ithaca, NY 14853, USA and
Materials Research Laboratory, Pennsylvania State University, University Park, PA 16802, USA

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An experimental method is proposed for the measurement of the photon's fundamental magnetic field operator $\hat{B}_{\Pi}$. It is shown that the quantum field and semi-classical descriptions of the optical Zeeman effect produce a different pattern of splitting in a $^1S \rightarrow {}^1P$ transition in an atom. The quantum field operator $\hat{B}_{\Pi}$ of the photon produces a pattern of three lines, each displaced from the original $^1S \rightarrow {}^1P$ line, while its classical equivalent $\boldsymbol{B}_{\Pi}$ produces a pattern akin to conventional Zeeman splitting with a static magnetic field.

Introduction

Recently, it has been deduced theoretically [1–4] that the photon generates a fundamental quantum of static magnetic flux density defined by the operator equation

$$\hat{B}_{\Pi} = B_0 \frac{\hat{J}}{\hbar} \tag{1}$$

where $\hat{J}$ is the photon's angular momentum operator, in the usual units of $\hbar$, the reduced Planck constant, and B_0 is the scalar magnetic flux density amplitude of a beam of light (N photons, for example a circularly polarised laser). The classical equivalent of the quantum field operator $\hat{B}_{\Pi}$ is an axial vector:

$$\boldsymbol{B}_{\Pi} = \pm B_0 \boldsymbol{k} , \tag{2}$$

where $\boldsymbol{k}$ is a unit axial vector in the propagation axis of the laser and where the $\pm$ signifies that $\boldsymbol{B}_{\Pi}$ changes sign with the laser's circular polarisation. It is emphasises at the outset that $\hat{B}_{\Pi}$ (or $\boldsymbol{B}_{\Pi}$) is *independent of frequency*, i.e. of the phase of the electromagnetic plane wave, and should not be confused with the usual magnetic field $\boldsymbol{B}$ of the plane wave, which is the usual oscillating field [5]. It is also important to note that the fundamental symmetries of $\hat{B}_{\Pi}$ (or $\boldsymbol{B}_{\Pi}$) are as follows. It is negative to the motion reversal operator $\hat{T}$, positive to the parity inversion operator $\hat{P}$, and evidently, from eq. (1), has the same $\hat{T}$ and $\hat{P}$ symmetries as the angular momentum operator $\hat{J}$. Thus $\hat{B}_{\Pi}$ (or $\boldsymbol{B}_{\Pi}$) is a fundamentally new concept in both quantum field theory and classical electrodynamic theory. It is a static, frequency independent, magnetic flux density, and eq. (1) shows that the photon generates $\hat{B}_{\Pi}$ at a fundamental level. It is well known, furthermore [6], that the photon, as a concept, is different from that of a particle such as an electron, proton, or neutron, because the photon is (a) massless, (b) propagates in vacuo at the speed of light, (c) *is not localised in space*. These properties are all contained succinctly in eq. (1), the first two through the well known properties of $\hat{J}$

Correspondence to: Prof. M.W. Evans, Department of Physics, University of North Carolina, Charlotte, NC 28223, USA.

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of the photon [5,6], the third through the factor B_0, which is the usual scalar magnetic flux density amplitude of a light beam. Clearly if this beam were to consist of only one photon, B_0 would remain finite and unlocalised in space. Shore [6] has recently provided an interesting discussion on the fundamental nature of the photon. It cannot be safely visualised as a massive particle [6], i.e. a localised concentration of energy and momentum, because it is massless and travels at the speed of light, so that there is no reference frame in which it can be viewed at rest [6]. Normal mode analysis argues against photons being localised in space, and the photon has no spatially localised wavefunction. Furthermore, plane wave photons are generated mathematically only by boundary conditions imposed on Cartesian coordinates, and as such are mathematical constructs, which nevertheless have a physical existence. The scalar B_0 is not spatially localised, because the beam intensity in watts per unit area depends on the square of B_0, and as a beam is focused, B_0 is obviously increased because the intensity is increased. In other words, B_0 is defined with respect to an area which is always finite. B_0 also appears in the classical equation (2), where $\boldsymbol{k}$ must not be confused with the propagation vector $\boldsymbol{\kappa}$. The latter is a $\hat{P}$ and $\hat{T}$ negative polar vector, and $\boldsymbol{k}$ is a P positive, T negative *axial unit vector*.

With these preliminaries eqs. (1) and (2) imply that a circularly polarised electromagnetic plane wave acts as a magnet, and it follows immediately that whenever a magnetic field is used experimentally, then, in principle, so can $\hat{\boldsymbol{B}}_\Pi$ (or $\boldsymbol{B}_\Pi$). Thus, it is possible to conceive of an optical Faraday effect, optical Zeeman effect (this work), an optical Stern–Gerlach experiment, optical NMR and optical ESR, optically induced bulk magnetization experiments, to name a few phenomena at random. To date, these optically induced magnetic effects have been approached with sporadically developed semi-classical theory, and the experimental evidence is fragmentary, being restricted to only two series of experiments spanned by some thirty years. The first of these was carried out by Pershan et al. [7–9] and by Shen [10], who used pulses from an early circularly polarised giant ruby laser to show that it could magnetize diamagnetic and a paramagnetic samples of liquids and doped glasses. The effect was named the 'inverse Faraday effect' (IFE), whose semi-classical description was developed by Kielich [11], Atkins and Miller [12], and by Manakov et al. [13] in terms of a light induced magnetic dipole moment mediated by a hypersusceptibility (magnetic/electric/electric), a mechanism which is to second order in the classical $\boldsymbol{B}_\Pi$ and whose magnetization can be expressed simply [14] as

$$|\boldsymbol{M}| = \tfrac{1}{3}|{}^{\mathrm{m}}\gamma^{\mathrm{ee}}|E_0^2 = \tfrac{1}{3}|{}^{\mathrm{m}}\gamma^{\mathrm{ee}}|c^2B_0^2$$
$$= \tfrac{1}{3}|{}^{\mathrm{m}}\gamma^{\mathrm{ee}}|(|\boldsymbol{B}_\Pi|)^2 . \tag{3}$$

Here $|{}^{\mathrm{m}}\gamma^{\mathrm{ee}}|$ denotes the magnitude of the tensorial hypersusceptibility term ${}^{\mathrm{m}}\gamma^{\mathrm{ee}}$ written out in full in ref. [14]; E_0 is the scalar electric field strength amplitude of an intense, circularly polarised laser and c is the speed of light. The notation $|\boldsymbol{B}_\Pi|$ means 'the (scalar) magnitude of the classical $\boldsymbol{B}_\Pi$ vector':

$$|\boldsymbol{B}_\Pi| = B_0 = \frac{E_0}{c} . \tag{4}$$

The theory of the IFE summarised in eq. (3) omits a term linear in $|\boldsymbol{B}_\Pi|$ which has recently been shown [15] to be a consequence of the existence of the classical $\boldsymbol{B}_\Pi$ field. In general, therefore, the semi-classical theory of the IFE produces

$$|\boldsymbol{M}| = A|\boldsymbol{B}_\Pi| + B(|\boldsymbol{B}_\Pi|)^2 + \mathrm{O}(|\boldsymbol{B}_\Pi|^3) \tag{5}$$

where A and B are coefficients defined by molecular property tensors, respectively susceptibility and hypersusceptibility. There is no reason to assume that the right hand side of eq. (5) is restricted to the term in $(|\boldsymbol{B}_\Pi|)^2$.

The second series of experiments, carried out recently by Warren et al. [16], has demonstrated the ability of circularly polarised, off-resonance, visible argon ion laser radiation to shift magnetic resonance frequencies in the enantiomers and racemic mixture of p-methoxyphenyliminocamphor, in its various solvents, and in deuteriated chloroform., This important demonstration followed some semi-classical predictions by the present author [17–20] and provides the first evidence for optical NMR (ONMR), or 'laser enhanced NMR spectroscopy' (LENS). If developed further, LENS promises well as a practical analytical technique, because for very modest laser intensities of up to about 3 watts per square centimetre [16], laser induced NMR shifts are small, *but measurable and site specific*, i.e. measurably different for each resonance site. It is clear that $\hat{B}_\Pi$ (or $\boldsymbol{B}_\Pi$) can play a direct and basic part in the theory of ONMR (and OESR [21]) because of the presence of interaction hamiltonians such as the first order terms

$$\Delta\hat{H}_1 = -\hat{m}_N \cdot \hat{B}_\Pi \; ; \tag{6}$$

$$\Delta\hat{H}_2 = -\hat{m}_N \cdot \boldsymbol{B}_\Pi \; . \tag{7}$$

Here $\hat{m}_N$ denotes a nuclear (or electronic) magnetic dipole moment operator for a given resonating entity such as a ^{1}H nucleus. In eq. (6) the hamiltonian operator $\Delta\hat{H}_1$ has been assumed to be a product of two *operators* (quantum field theory) and in eq. (7) $\Delta\hat{H}_2$ has been formed from the product of one operator and the classical field $\boldsymbol{B}_\Pi$, which is a vector (semi-classical theory).

It is not yet clear which of these hamiltonians is the more appropriate for ONMR (or OESR), but intuitively eq. (6) is expected to be more accurate, because there appears to be no reason to assume that $\hat{B}_\Pi$ acts classically when it is known from eq. (1) to be a fully quantised operator. More work is needed to interpret optical NMR and optical ESR theoretically, and more data are needed on model systems, but LENS is a technique which can be applied already in the analytical laboratory and the spectra interpreted as a fingerprint for a given sample. This procedure clearly helps to characterise and identify a sample supplemented by new information produced by an applied circularly polarised laser. There are several new useful variable parameters, for example laser intensity, frequency and polarity.

In terms of fundamental science, however, it is important to devise experimental means of measuring $\hat{B}_\Pi$. In this paper we propose a test through the optical Zeeman effect in singlet atom states, using $^1S \rightarrow {}^1P$ transitions split by a circularly polarised laser and measured spectroscopically with a probe beam in a high resolution spectrometer.

In section 1 the quantum field interaction energy is developed to give a pattern of optical Zeeman effect splitting of the atomic $^1S \rightarrow {}^1P$ line at visible frequencies, and it is shown that the result is three lines, each shifted from the original absorption frequency, one to higher frequency and two to lower frequency of the original line.

In section 2, the same procedure is carried out for a semi-classical interaction energy and the result is a different splitting pattern, consisting of three lines, a central undisplaced line flanked by two lines, displaced by the same amount to high and low frequency respectively.

A discussion of these theoretical findings is given, with order of magnitude estimates of the splitting for a given laser intensity.

1. The quantum field theory of the optical Zeeman effect

In this development we consider the splitting of an atomic $^1S \rightarrow {}^1P$ optical frequency absorption line by a circularly polarised laser by assuming that the interaction energy between this pump laser and the atom is the expectation value

$$\Delta E_\Pi = -\langle LJFM_F | \hat{m} \cdot \hat{B}_\Pi | L'J'F'M'_F \rangle \; . \tag{8}$$

Here $\hat{m}$ is the magnetic dipole moment operator of the atom, proportional to an orbital angular momentum operator $\hat{L}$:

$$\hat{m} = \gamma_e \hat{L} \tag{9}$$

through the gyromagnetic ratio γ_e, as usual. Associated with operator $\hat{L}$ is the angular momentum quantum number L. The $\hat{B}_\Pi$ operator is generated by the circularly polarised pump laser and defined by eq. (1) in terms of the photon's angular momentum operator $\hat{J}$ with which is associated the angular momentum quantum number J. The quantum number F in eq. (8) is defined by the usual Clebsch–Gordan conditions

$$F = L + J, \ldots, |L - J|; \qquad M_F = M_L + M_J . \tag{10}$$

It is immediately possible to develop this energy in terms of standard angular momentum coupling theory in quantum mechanics [22–24] to give the textbook result

$$\begin{aligned} &\langle LJFM_F|\hat{m}\cdot\hat{B}_\Pi|L'J'F'M_F'\rangle \\ &\quad = \delta_{FF'}\delta_{MM'}(-1)^{J+L'+F} \\ &\qquad \times \begin{bmatrix} L & J & F \\ J' & L' & 1 \end{bmatrix} \langle L\|\hat{m}\|L'\rangle\langle J\|\hat{B}_\Pi\|J'\rangle ; \\ &\hat{m} = \gamma_e\hat{L} ; \qquad \hat{B}_\Pi = B_0\frac{\hat{J}}{\hbar} . \end{aligned} \tag{11}$$

Here the braces denote the well-known 6-j symbol [22–24], which premultiplies reduced matrix elements as usual. This is recognisable as an entirely standard reduction of a spin–spin hamiltonian, whose expectation value is eq. (11), the interaction energy. The spin–spin content is given by the angular momentum operator product $\hat{L}\cdot\hat{J}$, a product of two 'spins'.

The expression (11) can be simplified considerably by restricting consideration to the diagonal matrix elements, again a standard textbook procedure. Before proceeding we note that the well-known triangle conditions [22] on the 6-j symbol in eq. (11) produce the selection rules

$$\Delta J = 0, \pm 1 ; \qquad \Delta L = 0, \pm 1 . \tag{12}$$

In general, however, there is no restriction from the triangle conditions on ΔF, provided that parity selection rules are obeyed. In our case this is always so because we consider transitions from ^{1}S states to singlet ^{1}P states of an atom. This is important in arriving at the nature of the optical Zeeman splitting in quantum field theory.

The diagonal components of eq. (11) can be reduced to a simple algebraic expression using the tabulated results [24]

$$\begin{aligned} \begin{bmatrix} L & J & F \\ J & L & 1 \end{bmatrix} &= -(1)^{J+L+F} \\ &\times \frac{F(F+1) - J(J+1) - L(L+1)}{(J(2J+1)(2J+2)L(2L+1)(2L+2))^{1/2}} \end{aligned} \tag{13}$$

and

$$\langle L\|\hat{L}\|L\rangle = (L(L+1)(2L+1))^{1/2}\hbar , \tag{14}$$

$$\langle J\|\hat{J}\|J\rangle = (J(J+1)(2J+1))^{1/2}\hbar , \tag{15}$$

giving the interaction energy

$$\begin{aligned} \Delta E_\Pi = -\tfrac{1}{2}(F(F+1) - J(J+1) \\ - L(L+1))\gamma_e B_0\hbar \end{aligned} \tag{16}$$

in a simple form. The Landé factor

$$g_L = \tfrac{1}{2}(F(F+1) - J(J+1) - L(L+1)) \tag{17}$$

is recognisable as that given by a simple vector coupling model [22–24] of L and J of the atom's $\hat{m}$ operator and the photon $\hat{B}_\Pi$ operator.

In the ^{1}S ground state we have $L = 0$, and we assume that $J = 1$ for the photon, although there is no a priori reason [5] to assume that a photon's J cannot exceed unity. Therefore $F = 1$ in the ^{1}S state. In the ^{1}P state, $L = 1$, $J = 1$ and $F = 2$, 1 and 0 from eq. (10). There are no restrictions on ΔF from the triangle rules on the existence of the 6-j symbol [22] so that transitions can occur between the $F = 1$ ground state ^{1}S and the three F states of ^{1}P. Therefore we observe three lines in the optical Zeeman spectrum from the quantum field interaction hamiltonian between operator $\hat{m}$ and operator $\hat{B}_\Pi$. In the ^{1}P state the original energy is split into three different components, with the original energy augmented by the three energies

$$\begin{aligned} \Delta E_\Pi(F=0) &= 2\gamma_e B_0\hbar , \\ \Delta E_\Pi(F=1) &= \gamma_e B_0\hbar , \\ \Delta E_\Pi(F=2) &= -\gamma_e B_0\hbar , \end{aligned} \tag{18}$$

causing the original ^{1}S-to-^{1}P frequency to be split into three, one line displaced by $-\gamma_e B_0/2\pi$ hertz to the high frequency side of the original, one displaced by $\gamma_e B_0/2\pi$ hertz to the low frequency side, and the third displaced by $2\gamma_e B_0/2\pi$ hertz to the low frequency side.

Therefore, this is the expected pattern of splitting from the $\hat{B}_\Pi$ operator of eq. (1) in the optical Zeeman effect under consideration. Note that in deriving this pattern the coupled representation of the angular momentum states has been used. In the Appendix we treat the same problem with an uncoupled representation, and show that in this case, the result obtained from quantum field theory is the same as that from the semi-classical theory, described in the following section.

2. Semi-classical theory of the optical Zeeman effect

In the semi-classical approach to the same spectrum as dealt with in section 1, the operator $\hat{B}_\Pi$ is replaced by the vector $\boldsymbol{B}_\Pi$. The interaction energy becomes

$$\Delta E_{\Pi,\mathrm{sc}} = -\langle LM_L|\hat{m}|L'M_L'\rangle \cdot \boldsymbol{B}_\Pi \tag{19}$$

because $\boldsymbol{B}_\Pi$ is no longer considered to be a quantum mechanical operator. The ^{1}P state of the atom is now augmented by the three energies

$$\begin{aligned} &\Delta E_{\Pi,\mathrm{sc}}(M_L = 1) = -\gamma_e \hbar B_0 , \\ &\Delta E_{\Pi,\mathrm{sc}}(M_L = 0) = 0 , \\ &\Delta E_{\Pi,\mathrm{sc}}(M_L = -1) = +\gamma_e \hbar B_0 , \end{aligned} \tag{20}$$

so that the original $^1\mathrm{S} \rightarrow {}^1\mathrm{P}$ absorption line is split into three lines, one of which is displaced by $-\gamma_e B_0/2\pi$ hertz to the high frequency side, one undisplaced line ($M_L = 0$) and one line displaced by $\gamma_e B_0/2\pi$ hertz to the low frequency side.

This splitting pattern is immediately recognisable as being the same as in the ordinary Zeeman effect [5] when there is no electronic spin angular momentum.

3. Discussion

The theoretical existence of the novel $\hat{B}_\Pi$ operator leads to a Zeeman pattern worked out in a simple atomic model in section 1. This pattern is found to be different from that expected from the classical $\boldsymbol{B}_\Pi$, worked out in section 2. We arrive at the interesting conclusion that the optical Zeeman effect is *different* in quantum field theory and in semi-classical theory. If verified experimentally this would appear to be unprecedented information on the flux quantum $\hat{B}_\Pi$.

The order of magnitude of the laser induced shift in both quantum and semi-classical approaches is estimated straightforwardly from the intensity (I_0) of the circularly polarised pump laser producing the operator $\hat{B}_\Pi$ [1–4]:

$$B_0 = |\hat{B}_\Pi| \sim 10^{-7} I_0^{1/2} , \tag{21}$$

and for a pump laser of intensity 100 watts cm^{-2} (10^6 watts m^{-2}), the shift $-\gamma_e B_0/2\pi$ is about 2 MHz. With contemporary high resolution spectroscopic techniques, such as double resonance, this is easily measurable. Clearly, the different theoretical pattern of optical Zeeman shifting from the quantum and semi-classical theories can be tested experimentally as a function of the laser intensity and circular polarity. Changing the circular polarity of the pump laser from right to left should change the sign of $\hat{B}_\Pi$ (or $\boldsymbol{B}_\Pi$) and if a linearly polarised pump laser is used, there should be no optical Zeeman splitting in either the quantum or semi-classical approach.

More generally, there should also be an anomalous optical Zeeman effect and an optical Paschen–Back effect, which occur when there is net electronic spin angular momentum in the atom as in a triplet state. We have considered the very simplest pattern of optical Zeeman splitting for clarity of exposition, and in general the optical Zeeman pattern is expected to be as rich in detail and analytical information as the well known conventional Zeeman effect, produced by a conventional magnet. In a sense, we have thus replaced the ordinary magnet by a 'light magnet', i.e. a magnet produced by a circularly polarised light beam.

Finally we consider the order of magnitude of second order effects which might interfere with the simple first order interaction energies (8) and (19). The nature of these second order effects has been investigated in full detail for atoms by Manakov et al. [13]. There are optical Stark effects, to second order in E_0, and therefore in B_0, and the second order optical Zeeman effect [13,25–28]. The latter occurs only with a circularly polarised pump laser and the second order optical Stark effects occur for both linearly and circularly polarised pump lasers, i.e. the optical Stark effect as considered by Manakov et al., for example, does not depend on the pump laser's circular polarity. Circular polarisation is essential, however, for the second order optical Zeeman effect. (There is no first order optical Stark effect for any polarisation.) An order-of-magnitude estimate of the likely shifts to second order can be made quite simply in the semi-classical approximation

$$\Delta E_{\Pi,\mathrm{sc}} = -\langle \hat{m} \rangle \cdot \boldsymbol{B}_{\Pi} + |\langle \boldsymbol{\alpha}'' \rangle| E_0^2 \tag{22}$$

where we have restricted consideration to the second order optical Zeeman term, mediated by the vectorial (i.e. the antisymmetric) atomic polarisability $\boldsymbol{\alpha}''$ [13,25–28]. Manakov et al. [13] have computed this ab initio for some atomic states and it is of the order 10^{-42} $\mathrm{C^2m^2J^{-1}}$. With the fundamental electrodynamic relation

$$I_0 = \tfrac{1}{2}\varepsilon_0 c E_0^2 \tag{23}$$

we find for a modest circularly polarised pump laser intensity I_0 of 1.0 watt $\mathrm{cm^{-2}}$ that the first order effect in eqs. (8) or (19) dominates by roughly six orders of magnitude, and the first order effect (linear in $\hat{B}_{\Pi}$ or $\boldsymbol{B}_{\Pi}$) continues to dominate when the pump laser's intensity is pulsed to about 10^{10} watt $\mathrm{m^{-2}}$.

4. Conclusion

The optical Zeeman effect is one consequence of the theoretical existence of the new flux quantum $\hat{B}_{\Pi}$ of electromagnetic radiation, and it has been demonstrated that the expected splitting is different in quantum field theory and semi-classical theory. The order of magnitude of the shift for modest pump laser intensity is also easily measurable in principle.

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Appendix

The uncoupled representation in quantum field theory

The quantum field theory applied in section 1 uses the *coupled* representation [5] of angular momentum states of the operators $\hat{L}$ and $\hat{J}$. In the coupled representation, the total angular momentum has a well defined magnitude and a well defined component M_F in the Z axis (the axis of propagation of the pump laser), but the individual projections of $\hat{L}$ and $\hat{J}$ on the Z axis are indeterminate: all that is known is that their sum must be M_F.

In the *uncoupled* representation of the same problem, the projections M_L and M_J are *both* specified, but there is no information about the relative orientation [5] of $\hat{J}$ and $\hat{L}$, and the total angular momentum is indeterminate. In this case the relevant Schrödinger equation is

$$\begin{aligned} &-\hat{m} \cdot \hat{B}_{\Pi} |LM_L;\, JM_J\rangle \\ &\quad = -\hbar M_L M_J \gamma_e B_0 |LM_L;\, JM_J\rangle \,, \end{aligned} \tag{A.1}$$

i.e. the eigenvalue of the operator product $-\hat{m} \cdot \hat{B}_{\Pi}$ is defined through the M_L and M_J azimuthal quantum numbers. The expectation value of the hamiltonian is therefore the energy change

$$\Delta E_{\Pi} = -\hbar M_L M_J \gamma_e B_0 \quad \text{(A.2)}$$

due to the applied circularly polarised pump laser. For a given circular polarisation M_J is either 1 or -1 [5]. Taking the value

$$M_J = 1\,, \quad \text{(A.3)}$$

the energy change in the ^1S state is zero (because $L = 0$, $M_L = 0$), but in the ^1P state there are three possible energy changes, corresponding to $L = 1$, $M_L = -1, 0, 1$:

$$\begin{aligned} &\Delta E_{\Pi}(M_L = -1) = \hbar\gamma_e B_0\,, \\ &\Delta E_{\Pi}(M_L = 0) = 0\,, \\ &\Delta E_{\Pi}(M_L = 1) = -\hbar\gamma_e B_0\,. \end{aligned} \quad \text{(A.4)}$$

Transitions are allowed between the single ^1S state and the three ^1P states according to the selection rule [5]

$$\Delta M_L \text{ (from } {}^1\text{S to } {}^1\text{P)} = \begin{cases} 0 & \text{(pi line)}\,, \\ \pm 1 & \text{(sigma lines)}\,, \end{cases} \quad \text{(A.5)}$$

and the original line is split into three: one line at the original frequency, flanked to high frequency by a line displaced by $-\gamma_e B_0/2\pi$ hertz from the original, and to low frequency by a line displaced by $+\gamma_e B_0/2\pi$ hertz.

This is recognisable as the same pattern as obtained in the well-known conventional Zeeman splitting [5] of the $^1S \rightarrow {}^1P$ line of an atom by a conventional magnetic field.

In summary, a 'conventional' Zeeman splitting pattern is obtained in quantum field theory in the *uncoupled* representation, and a novel pattern of splitting in the coupled representation of section 1 of the text. Presumably, the uncoupled representation is appropriate in a very intense laser field, and we call this the 'optical Paschen–Back Effect'.

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