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Gain without inversion: why and how?

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Lasing without inversion: a useful concept?

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The historical development that led to the modern concept of amplification and lasing without inversion is reviewed. The two basic mechanisms for amplification without inversion and their theoretical formulation are discussed. Finally, the three experiments that confirm the occurrence of these effects are described.

1. Introduction

1.1. Early history

In the past few years, there has been much activity on the subject of lasing without inversion (LWI) and/or amplification without inversion (AWI), although most workers disagree on the precise definition on what they mean by LWI or AWI. Therefore, let me try to define the topic first. Taking a simple and naive approach, a laser operating with a two-level medium will start to lase if there is sufficient population inversion between the two levels so that the gain can balance the cavity losses. Let N_1 and N_2 be the populations of the two lasing levels and $E_1 = \hbar\omega_1$ and $E_2 = \hbar\omega_2$ their corresponding energy. The lasing threshold can be expressed as

$$N_2 - N_1 > N_{th} > 0, \quad (1)$$

where N_{th} is a positive constant that depends linearly on the cavity losses. The point that should be stressed at this stage is that N_2 and N_1 are the populations of the two levels in the absence of interaction with the field, but taking fully into account the effect of all incoherent optical pumping processes and the finite lifetime of the atoms. Equation (1) expresses the fact that amplification requires an inversion of population. The difficulty with this scheme is that, in order to generate a coherent field at the frequency $\omega_2 - \omega_1$, it is necessary to have an incoherent pumping at a higher frequency since both levels E_1 and E_2 are excited levels. In other words, a conventional laser transforms incoherent energy into a coherent field at a lower frequency.

A more sophisticated laser theory confirms that equation (1) is indeed a necessary condition for lasing if we only consider incoherent optical pumping and

one-photon interactions and wish to amplify a field which is resonant with the two-level energy difference: $\omega_{\text{field}} = \omega_2 - \omega_1$. However, more elaborate schemes are possible. The simplest of them is based on the phenomenon of Rabi oscillations. When a two-level atom interacts with a periodic electric field, Rabi has shown that the probability of finding the electron in either of the two levels varies periodically between zero and 1. The frequency of this transfer is the Rabi frequency $\Omega = \mathcal{P}\mathcal{E}/\hbar$ which is proportional to the driving field amplitude \mathcal{E} and to the matrix element \mathcal{P} of the dipole moment between the two states. In the optical domain, the fields that are used in laboratories verify the inequality $\omega_2 - \omega_1 \gg \Omega$. In a lasing cavity, the nonlinear interaction between the resonant field oscillating at the atomic frequency $\omega_2 - \omega_1$ and the field-induced population oscillation at the Rabi frequency results in the occurrence of beat notes $\omega_2 - \omega_1 \pm \Omega$, known as the Rabi sidebands. It has been predicted by Rautian and Sobel'man (1961) and verified experimentally (Wu *et al.* 1977, Grandclément *et al.* 1987, Ze'likovich *et al.* 1987, Khitrova *et al.* 1988, Lezama *et al.* 1990) that, if a two-level medium is driven by a strong resonant coherent electric field, amplification without population inversion ($N_1 > N_2$) is possible at the Rabi side band. Thus gain is possible away from the atomic resonance in the absence of population. This means a transfer of coherence from one field to another field. However, this transfer is realized without extraction of energy from the medium. The theoretical interpretation of this scheme has been given by Cohen-Tannoudji and Reynaud (1977) and Knight and Milonni (1980) in terms of multiphoton processes and inversion between dressed states. The difficulty with this scheme is that the frequency up-conversion which is realized depends in a sensitive way on the driving field amplitude which is prone to intrinsic fluctuations. Thus, the corresponding field

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frequency will be hard to stabilize. Furthermore, arbitrary field frequencies require arbitrarily powerful lasers, a practical impossibility. Clearly, the situation would be much better if the amplified field frequency could be tied to some stable property such as energy levels. The same critical comment is valid for the early suggestions of AWI made by Marcuse (1963) and Holt (1976) who based their mechanism on the Doppler recoil splitting of the emission and absorption spectra.

1.2. Atomic interference

Although it is difficult to trace back the first application of this concept in cavity nonlinear optics, early references that explicitly mention interference as a mechanism to manipulate atomic transition rates are those of Popova *et al.* (1970a,b). A classic review on coherent trapping is the report of Hoo and Eberly (1985). To understand the principle of atomic interference, we consider a three-level medium (figure 1). It is important to realize that there is no three-level medium in nature. What we really mean is that only three levels interact via coherent fields in this scheme, in addition to (at least) a fourth level which serves as a reservoir of atoms. The presence of the other levels is essential to escape from contradictions that would otherwise arise if there were really only three levels. These contradictions are related to the selection rules which forbid, for instance, that all transitions be allowed in a three-level medium (although for sufficiently asymmetric molecules, this rule may be broken).

Figure 1 describes a situation in which two monochromatic fields couple the upper level 3 to the

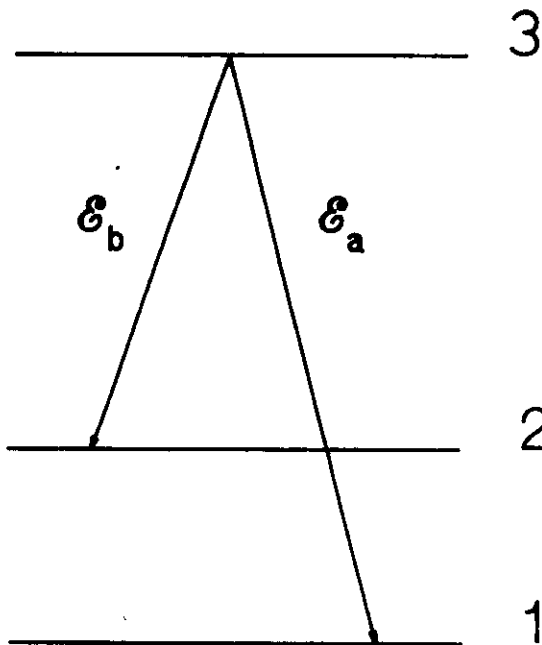


Figure 1. Three-level configuration and the A-scheme.

lower levels 1 and 2. The total electric field is

$$E(t) = \mathcal{E}_a \cos(\omega_a t) + \mathcal{E}_b \cos(\omega_b t), \quad (2)$$

and the atomic wavefunctions are $\{\varphi_j, j=1, 2, 3\}$. Suppose that we can create a superposition state involving the two lower states

$$\begin{aligned} \varphi_s &= c_1 \varphi_1 + c_2 \varphi_2 \\ &= C_1 \varphi_1 \exp(-i\omega_1 t) + C_2 \varphi_2 \exp(-i\omega_2 t) \end{aligned} \quad (3)$$

Then the transition probability between this superposition state and the upper state 3 is

$$W = |\langle \varphi_3 | -er \cdot \varepsilon E(t) | \varphi_s \rangle|^2, \quad (4)$$

where $-er$ is the electric dipole excited by the electric field whose polarization is ε . To proceed further, we need to specify the selection rules. If each of the two monochromatic fields \mathcal{E}_a and \mathcal{E}_b can couple the upper state to either of the lower states, then equation (4) becomes

$$\begin{aligned} W_1 &= \frac{1}{4} \mathcal{P}^2 \mathcal{E}^2 |C_1 [1 + \exp(-i\omega_{21} t)] \\ &\quad + C_2 [1 + \exp(i\omega_{21} t)]|^2. \end{aligned} \quad (5)$$

For simplicity, we have assumed that

- (1) all non-vanishing matrix elements are equal to $\mathcal{P}\mathcal{E}$;
- (2) variations in the electric field over an atomic wavelength are negligible; and
- (3) the field frequencies fulfil the condition of relative resonance

$$\omega_{31} - \omega_a = \omega_{32} - \omega_b, \quad (6)$$

with the notation $\omega_{pq} \equiv \omega_p - \omega_q$.

The transition probability W_1 is a periodic function of time which oscillates at the low-frequency separation between the two lower levels. However, a more interesting situation occurs if the selection rules are that each field component couples the upper level to only one of the lower levels. Then the transition probability (4) becomes

$$W_1 = \frac{1}{4} \mathcal{P}^2 \mathcal{E}^2 |C_1 + C_2|^2. \quad (7)$$

There exists an interesting state corresponding to the choice $C_1 = -C_2$. We shall call such a state a *trap state*. It is characterized by the properties

$$\Psi_{\text{trap}} \propto \varphi_1 - \varphi_2, \quad |\langle \Psi_{\text{trap}} | -er \cdot \varepsilon E | \varphi_3 \rangle|^2 = 0. \quad (8)$$

Thus, any atom which is prepared in the trap state will be unable to interact with the upper state. As is obvious from the above discussion, this effect results from the *interference of atomic wavefunctions* between the two excitation mechanisms; the coupling of levels 3 to 1 via the field \mathcal{E}_a and the competing coupling of levels 3 to 2

via the field \mathcal{E} , interfere destructively. A description of this interference requires a quantum description of the atoms, although the field can be treated classically. This is precisely the framework of the semiclassical formulation of nonlinear optics and laser physics, which we shall use later in this paper.

Independently of these speculations, Alzetta *et al.* (1976) from Pisa reported the first experimental evidence of atomic interference in agreement with the above simple model. The principle of the Pisa experiment is simple. A cylindrical cell contains sodium vapour in a suitable buffer gas. Along the longitudinal axis, z , a static magnetic field is applied to create a Zeeman splitting. The magnetic field increases linearly along the cell axis. Therefore, the Zeeman splitting is also a linear function of z . A multimode laser beam is sent into the cell in the z direction and the fluorescence light is observed perpendicular to the cell axis. An essential ingredient in the experiment is that the laser operates in the locked regime; all modes are interdependent in such a way that their frequency differences are locked, that is, remain constant. The experimental finding is that the fluorescence spectrum displays dark lines at specific values of z . A dark line means that no atoms can emit in the upper state, that is, there is no transition from the upper state to a lower state. A careful analysis of this experiment indicates that the values of z at which dark lines occur are such that the Zeeman splitting equals an integer multiple of the laser intermode frequency. This is just the condition of relative resonance (6) which was used in the analysis of the interference process, and indeed the concept of atomic coherence provides a framework in which the Pisa experiment finds a natural and simple explanation as shown by Arimondo and Orriols (1976). Independently of the Pisa group, Gray *et al.* (1978) also found dark lines but in the hyperfine structure of sodium and using a slightly different experimental set-up.

1.3. Fano interference

An elaboration on the interference concept applied to AWI was put forward by Arkhipkin and Heller (1983). They considered a two-level system with the upper level embedded in the continuum (i.e. the upper level lies above the lowest ionizing threshold). The principle at stake here is that interference between the resonant and the non-resonant channels of transitions to the continuum provides an asymmetry between the absorption and emission spectral profiles. These authors showed that this asymmetry may lead to AWI. The theory of transitions involving the continuum had been established by Fano (1961) and Fano and Cooper (1968) and was recently reviewed by Knight *et al.* (1990). Hel'or and Popov (1976) had suggested a way to induce

transitions of arbitrary range in the continuum, and experimental evidence (Heller *et al.* 1981, Dimov *et al.* 1983) provided some support of their theory. Experimentally, these transitions are realized with intense lasers that provide sufficient power to induce multiphoton transitions. However, it is difficult to control all parameters in these experiments: the conclusions of these early experiments remains a subject of debate.

1.4. Why amplification and lasing without inversion?

At this point, it is perhaps appropriate to discuss the reason(s) that lead physicists to pursue AWI and LWI. In the early papers, AWI and LWI appeared more as a byproduct of a theoretical analysis than as the purpose of the work. This soon changed. Why try to find schemes that lead to lasing without inversion? What is wrong with the usual schemes? The reason becomes obvious if we try to imagine a scenario that would produce an X-ray or even a γ -ray laser. It is rather difficult to create an inversion of population using incoherent pumping at or above these frequencies. Thus the road to higher-frequency lasers is not accessible via conventional incoherent pumping.

An alternative possibility would be to use the Raman process. Raman lasers are common in the infrared. They result from the coherent pumping by another laser of a medium which does not need a population inversion. The basic mechanism involves two-photon transitions, each photon inducing in general a non-resonant transition. Depending on the level configuration, the generated field will have a low or a high frequency (Stokes or anti-Stokes scattering, respectively). The difficulty with the Raman lasers is that they do not extract energy from the nonlinear medium: the medium serves only as a support to the scattering process that results in the transfer of coherence and energy from one field to the other field. This is especially apparent from the fact that a Raman laser can operate with no atoms in the upper state†.

With these remarks in mind, we can define AWI as a mechanism that will lead to the amplification of a weak probe field by transfer of energy from a strong and coherent driving field (if any) oscillating at a lower frequency than the probe field *and* by extraction of energy from the material medium without the prerequisite of a population inversion *between the lasing levels*. The populations to which we refer here are the 'initial' atomic populations, that is those reached when

† More exactly, in the case of Stokes scattering, no population inversion is required. In the anti-Stokes case a small amount of energy is extracted from the lower two levels (in a three-level scheme) and a population inversion between the two lower levels is required.

there is no interaction with the coherent fields although all incoherent decay and pumping process are in action.

For instance, in the case of the three-level medium in figure 1, we seek amplification with $N_1 > N_3$ and $N_2 > N_3$, the trick being that, if there are atoms in the trapped state, all that is necessary to achieve gain is that the number of atoms in the remaining two lower levels ($N_1 + N_2 - N_{\text{trap}}$) is less than the number of atoms in the upper level. In this way, an effective population inversion is obtained and energy can be extracted from the material medium in the usual way. This situation is usually referred to as hidden inversion.

1.5. Recent history

Despite the few papers cited in §1.2, the all-important concept of atomic interference was lost to most of the laser community although it remained an active subject of research in the field of multiphoton transitions. The field of AWI and LWI found its real start in two papers, published independently.

Kocharovskaya and Khanin (1986) had started to investigate systematically the interaction of three-level media with pulsed coherent monochromatic and steady polychromatic fields. They had found earlier that atoms could be trapped in a superposition state such as (8) if a three-level medium interacts with a train of laser pulses provided that, first, the pulse duration is much shorter than the period of the low-frequency oscillations $2\pi/(\omega_2 - \omega_1)$, and second, the pulse repetition frequency is a multiple of the low-frequency oscillations. This trapping leads to a reduced absorption coefficient of the medium. Finally, they realized (Kocharovskaya and Khanin 1988) that under suitable conditions the threshold for amplification could be reached in this scheme, leading to gain. This implies the excitation of the low-frequency coherence between the levels 1 and 2 in figure 1 in the medium via some external fields. It leads to a condition which takes the form of inequalities that must be verified by the various atomic decay rates and by the atomic populations. Following a completely different line of thought, Harris (1989) proposed an AWI scheme that could operate in the steady-state, where two upper states are above the ionizing threshold and which decay to the *same* continuum. This decay to a common continuum is the source of the interference that eventually leads to AWI. Very quickly, Scully *et al.* (1989) realized that the interference scheme of Harris could be obtained without recourse to the autoionizing states and the complex theory that they require. Rather, they showed in one of the most elegant papers in this domain that AWI can result from the interference of two excitation paths in the Λ -scheme in figure 1, when the low-frequency transition is driven by a microwave field

and the two probe fields have equal frequencies $\omega_3 - \frac{1}{2}(\omega_2 - \omega_1)$. Furthermore, their quantum analysis of the Λ -scheme could be 'inverted' and applied to the V-scheme. Here the prediction is that interference can lead to population inversion without lasing!

After these initial papers an explosive growth of theoretical work took place and resulted this year in the publication of three papers reporting clear experimental evidence of AWI, albeit in the pulsed or transient regime. These results will be described below.

2. Formulation

One of the problems with AWI and LWI is that there is a large gap between the simple description in §1.2 (which is essentially correct but oversimplified) and a more rigorous analysis. The lack of appreciation of this gap has led to some misunderstandings, conflicts and many wrong results. Since we have to deal with unstable atomic levels, there is no way, as of now, to derive a self-consistent theory from first principles; some admixture of phenomenology is unavoidable. Let us therefore proceed carefully with the derivation of a framework in which we can describe the interaction of multilevel atoms interacting with multicomponent coherent fields and stress those points which are not common with the theory of two-level systems.

2.1. Irreversible dynamics

We consider a collection of unstable atoms interacting with incoherent sources that affect the population of all levels. The natural way to describe quantized atoms is via the Schrödinger wavefunction. With each atom, we associate a wavefunction which is decomposed on an orthogonal basis $\Psi_a(t) = \sum_n C_a(n, t) \phi_a(n)$. However, what is observable are the bilinear products of the coefficients $C_a(n, t)$: the probabilities $C_a(n, t) C_a^*(n, t)$ and the coherences $C_a(n, t) C_a^*(n', t)$. Hence we introduce the density matrix $\rho(n, n', t) = \langle C_a(n, t) C_a^*(n', t) \rangle$ where the angular brackets indicate an average over all the atoms of the system. The Schrödinger equation implies the von Neumann equation for the density operator $i\hbar \partial \rho / \partial t = H\rho - \rho H$. The matrix elements of ρ are the $\rho(n, n', t)$. The reason for introducing the density matrix formulation is directly connected with the fact that we have to describe unstable atomic states. It is conceptually much simpler to introduce the phenomenological constants such as damping and pumping rates for the observable probabilities and coherences than for the wavefunction.

For stable atoms in the absence of interactions, we have from the Schrödinger equation $i\partial \rho_{pq} / \partial t = i(\omega_p - \omega_q) \rho_{pq}$; populations do not vary and coherences

oscillate. When the atomic lifetime is included, the off-diagonal matrix elements will display damped oscillations:

$$\left. \frac{\partial \rho_{pq}}{\partial t} \right|_{\text{irrev}} = [-\gamma_{pq} + i(\omega_p - \omega_q)] \rho_{pq}, \quad p \neq q. \quad (9)$$

For the irreversible atomic population dynamics, we follow the traditional formulation (see the excellent account in the book by Haken (1970):

$$\left. \frac{\partial \rho_{pp}}{\partial t} \right|_{\text{irrev}} = - \sum_p \rho_{pp} W(p \rightarrow q) + \sum_q \rho_{qq} W(q \rightarrow p), \quad (10)$$

where the sum is over all atomic levels. This equation expresses the fact that the population of level p is affected by two processes: depopulation of level p via transitions from level p to another level q and repopulation of level p via transitions from another level q to the level p . These processes are characterized by transition probabilities $W(p \rightarrow q)$. This transition probability must account for two properties. The transition $p \rightarrow q$ is possible only if there are atoms in level p . Hence $W(p \rightarrow q)$ must be proportional to N_p , the population of level p . The rate at which transitions occur in a macroscopic atomic sample is, by definition, the population decay time for that transition. We define this decay time by

$$W(p \rightarrow q) \equiv \frac{N_p}{T_1^{pq}}. \quad (11)$$

A property which we shall need but is not obvious to justify is that the population decay time T_1^{pq} equals T_1^{qp} . This comes from considerations of non-equilibrium statistical mechanics which are out of place here. With these results, we can write

$$\begin{aligned} \left. \frac{\partial \rho_{pp}}{\partial t} \right|_{\text{irrev}} &= - \sum_q \frac{\rho_{pp} N_q}{T_1^{pq}} + \sum_q \frac{\rho_{qq} N_p}{T_1^{qp}} \\ &= - \sum_{q \neq p} \frac{\rho_{pp} N_q - \rho_{qq} N_p}{T_1^{qp}} \equiv R_p. \end{aligned} \quad (12)$$

It is clear that the N_p introduced in the population dynamics are the variables in terms of which we wish to express the condition of amplification since in steady state the solution of equation (12) is $\rho_{pp} = N_p$. An essential result for the discussion of these amplification conditions is the relation between the polarization and the population decay rates. The polarization decay rate is defined through (Haken 1970)

$$\gamma_{pq} \equiv \frac{1}{T_2^{pq}} = \frac{1}{2} \sum_k [W(p \rightarrow k) + W(q \rightarrow k)]. \quad (13)$$

The sum over the states is not restricted and the $W(p \rightarrow q)$ are real positive coefficients that describe phase-destroying processes due to virtual transitions. They originate from diagonal higher-order process such

as $p \rightarrow q \rightarrow p$; they leave the populations invariant but modify the coherences. With this definition, the fundamental inequality relating the decay rates is easily derived:

$$\begin{aligned} 2\gamma_{pq} &= \sum_k [W(p \rightarrow k) + W(q \rightarrow k)] \geq \sum_{k \neq p} W(p \rightarrow k) \\ &\quad + \sum_{k \neq q} W(q \rightarrow k), \end{aligned} \quad (14)$$

or equivalently

$$\frac{2}{T_2^{pq}} \geq \sum_{k \neq p} \frac{N_k}{T_1^{pk}} + \sum_{k \neq q} \frac{N_k}{T_1^{qk}}. \quad (15)$$

This inequality establishes a relation between the decay rates and the atomic populations. For instance, increasing the population of the upper level in a three-level medium increases the decay rate between the remaining two levels and hence reduces the corresponding coherence. If we consider the degenerate case in which all population decay times are equal ($T_1^{pk} \equiv T_1$) and all coherence decay time are also equal ($T_2^{pk} \equiv T_2$), a direct consequence of equation (15) is that $2/T_2 \geq (1 + N_k)/T_1$. Therefore, we have the constraint $3/T_2 \geq 2/T_1$ instead of the two-level inequality $2/T_2 \geq 1/T_1$.

2.2. The complete dynamics

The reversible part of the dynamics results from the Hamiltonian equations which lead to the von Neumann equation

$$i\hbar \left. \frac{\partial \rho}{\partial t} \right|_{\text{rev}} = [H, \rho]. \quad (16)$$

By adding the irreversible dynamics to the Hamiltonian dynamics we obtain a description of the light-matter interaction. Since the crucial element that we intend to describe is the occurrence of atomic interference, a semiclassical theory of light-matter interaction, in which matter is quantized but the field is classical will be sufficient (Haken 1970, Sargent *et al.* 1974).

In this review, we shall consider only the AWI problem since very few papers deal with the LWI question and experimental results are still focused on the demonstration of gain in systems without inversion.

3. Theoretical developments

3.1. The simple A-scheme

We first consider the A-scheme in figure 1. To formulate the problem, we consider a medium whose active constituent are three-level atoms. They are homogeneously distributed in the sample and all atoms are equivalent (approximation of an Einstein solid). In this

medium, we launch a field composed of two monochromatic waves,

$$E_a = \mathcal{E}_a \exp(-i\omega_a t + ik_a z) + \mathcal{E}_a^* \exp(i\omega_a t - ik_a z)$$

and $E_b = \mathcal{E}_b \exp(-i\omega_b t + ik_b z) + \mathcal{E}_b^* \exp(i\omega_b t - ik_b z)$. The field E_a has a frequency ω_a close to $\omega_3 - \omega_1$ while the field E_b has a frequency ω_b close to $\omega_3 - \omega_2$. The medium is not bounded in the field propagation direction because all we seek is to determine the gain condition. Laser action would require the added complication of a resonant cavity and its nonlinear feedback. As the field propagates in the medium, it interacts with the atoms via dipole interactions proportional to the matrix element of the electric dipole between the states 3 and 1 for the field E_a and between the states 3 and 2 for the field E_b . These transitions induce the coherences and population variations according to the Hamiltonian part of the evolution as described by equation (16). The irreversible contribution is simply added to the Hamiltonian evolution. Since the \mathcal{E} are slowly varying amplitudes (variations on the scale of the optical frequency and wavenumber have already been factored out), we shall also deal with the slowly varying envelopes of the coherences: $\sigma_{31} = \rho_{31} \exp(i\omega_a t)$, $\sigma_{32} = \rho_{32} \exp(i\omega_b t)$ and $\sigma_{21} = \rho_{21} \exp[i(\omega_a - \omega_b)t]$. For the fields, we use Maxwell equations and take advantage of the fact that the amplitudes \mathcal{E} vary slowly in space and time to simplify the propagation equations. Eventually, this system is described by the following equations (Kocharovskaya and Mandel 1990):

$$\frac{\partial \alpha}{\partial t} + \frac{1}{c_a} \frac{\partial \alpha}{\partial z} + \kappa_a \alpha = \frac{2\pi i N \omega_a |\mu_{31}|^2 \sigma_{31}}{c_a \hbar}, \quad (17)$$

$$\frac{\partial \beta}{\partial t} + \frac{1}{c_b} \frac{\partial \beta}{\partial z} + \kappa_b \beta = \frac{2\pi i N \omega_b |\mu_{32}|^2 \sigma_{32}}{c_b \hbar}, \quad (18)$$

$$\frac{\partial \sigma_{31}}{\partial t} = -\sigma_{31}[\gamma_{31} + i(\omega_3 - \omega_1 + \omega_a)] + i(\alpha n_{13} + \beta \sigma_{21}), \quad (19)$$

$$\frac{\partial \sigma_{32}}{\partial t} = -\sigma_{32}[\gamma_{32} + i(\omega_3 - \omega_2 + \omega_b)] + i(\alpha \sigma_{12} + \beta n_{23}), \quad (20)$$

$$\frac{\partial \sigma_{21}}{\partial t} = -\sigma_{21}[\gamma_{21} + i(\omega_2 - \omega_1 + \omega_a - \omega_b)] + i(\beta^* \sigma_{31} - \alpha \sigma_{23}), \quad (21)$$

$$\frac{\partial \rho_{11}}{\partial t} = R_1 + 2 \operatorname{Im}(\alpha \sigma_{13}), \quad \frac{\partial \rho_{22}}{\partial t} = R_2 - 2 \operatorname{Im}(\beta \sigma_{23}), \quad (22)$$

where α and β are proportional to the fields \mathcal{E}_a and \mathcal{E}_b , respectively. The functions R_p are defined in equation (12) and $n_{pq} \equiv \rho_{pp} - \rho_{qq}$.

Equations (17) and (18) describe the propagation of the two fields in a medium where each field excites one atomic transition. Equations (19)–(21) describe the evolution of the atomic coherences or polarization under the influence of the field and the incoherent process. Finally, equation (22) describes the population dynamics (with $\operatorname{Tr}(\rho) = 1$) under the influence of both the field propagation in the medium and the incoherent processes. Since we are dealing with a propagation problem, it is natural to seek normal waves, that is, solutions of the type α and $\beta \propto \exp(-i\omega t + ikz)$. This assumption implies that σ_{31} and σ_{32} will also be proportional to $\exp(-i\omega t + ikz)$. Let us therefore define

$$\operatorname{col}(\alpha, \beta, \sigma_{31}, \sigma_{32}) = \operatorname{col}(a, b, s_{31}, s_{32}) \exp(-i\omega t + ikz), \quad (23)$$

where the notation $\operatorname{col}(a, b, \dots)$ stands for a column vector whose components are a, b, \dots . In equation (23), ω is real and k will be given by the dispersion relation $k = k(\omega)$ to be derived. After inserting this expression into equations (17)–(22), we find a set of amplitude equations. They admit a steady-state solution. Solving the algebraic material equations for the density matrix elements and inserting this result into the two field equations leads to two homogeneous equations for a and b : $A_{11}a + A_{12}b = 0$, $A_{21}a + A_{22}b = 0$. The compatibility condition $A_{11}A_{22} = A_{12}A_{21}$ is the dispersion relation $k = k(\omega)$. When this dispersion relation is verified, the solution can be written as an expression for the ratio of the two amplified fields. The dispersion relation is complex, leading to a complex wavenumber k . If $\operatorname{Im}(k) > 0$, the solution (23) will be damped as z increases; the field is progressively absorbed by the medium. However, if $\operatorname{Im}(k) < 0$, the solution (23) grows exponentially with z and the field is amplified. At line centre $\omega_3 - \omega_1 + \omega_a - \omega = \omega_3 - \omega_2 + \omega_b - \omega = 0$ and for negligible losses $\kappa_a = \kappa_b = 0$, the amplification condition takes the form

$$|\sigma_{21}|^2 \equiv |\rho_{21}|^2 > (\rho_{11} - \rho_{33})(\rho_{22} - \rho_{33}). \quad (24)$$

Thus the key role is played, in this scheme, by the low-frequency coherence σ_{21} . It is a simple exercise to verify that at threshold, that is, for $|\rho_{21}|^2 = (\rho_{11} - \rho_{33})(\rho_{22} - \rho_{33})$, we have $\sigma_{13} = \sigma_{23} = 0$ so that the solution of the eigenvalue problem $\rho\Psi = \Lambda\Psi$ is

$$\Lambda_1 = \rho_{33}, \quad \Psi_1 = -(\rho_{22} - \rho_{33})^{1/2} \varphi_1 + (\rho_{11} - \rho_{33})^{1/2} \varphi_2 \propto \beta \varphi_2 + \alpha \varphi_1, \quad (25)$$

$$\Lambda_2 = 1 - 2\rho_{33}, \quad \Psi_2 = +(\rho_{22} - \rho_{33})^{1/2} \varphi_1 + (\rho_{11} - \rho_{33})^{1/2} \varphi_2 \propto \alpha \varphi_2 - \beta \varphi_1, \quad (26)$$

$$\Lambda_3 = \rho_{33}, \quad \Psi_3 = \varphi_3. \quad (27)$$

where $\varphi_j = \text{col}(\delta_{1j}, \delta_{2j}, \delta_{3j})$ are the unperturbed atomic states. In the state Ψ_2 we recognize the generalization of the trap state introduced in section 1.2; there is indeed no transition between that state and the upper state Ψ_3 . The state Ψ_1 is the state complementary to the trap state; atoms prepared in Ψ_1 can interact with the upper state. However, at threshold, the eigenvalues indicate that both states are equally populated which imply that they are transparent to the radiation!

Two remarks are in order at this point. First, it has been shown by Kocharovskaya *et al.* (1991b) that the condition (24) is equivalent to the condition of population inversion between the states Ψ_3 and Ψ_1 . Second, and most important, the present analysis is a real case study of the dangers provided by a linear stability analysis. If we pay some attention to the results obtained so far, it is not very difficult to realize that, if amplification is achieved in this Λ -scheme, the problem of the *perpetuum mobile* is solved. It requires a nonlinear analysis to prove that the threshold (24) cannot be reached by finite-intensity beams; it is an asymptotic upper bound and the coherent fields cannot be amplified with fewer atoms in the upper state than in either of the two lower states. What remains of this analysis are two indications: first, AWI in the Λ -scheme requires an additional source of coherence; second, although gain cannot be achieved via this mechanism, the nonlinear analysis shows that the absorption can be drastically reduced. In some limiting case, it was shown (Kocharovskaya and Mandel 1990) that, in the steady regime, the field intensity satisfies a law of the type

$$\frac{dI}{dz} \propto -\frac{I}{1 + I(3/I_s + 1/I_c)}, \quad \frac{I_c}{I_s} = \gamma_{21} T_1^{32}. \quad (28)$$

I_s is the usual saturation intensity whereas I_c is the coherence intensity. It is quite possible to have $I_c/I_s \ll 1$ and $I_0 \gg I_c$, which leads to coherent bleaching (or electromagnetically induced transparency) at a low intensity, as opposed to saturation bleaching which would require a fairly large initial intensity $I_0 \gg I_s$.

3.2. The driven Λ -scheme

The conclusion of the previous section is that an additional source of coherence is required. With three levels, there is quite a number of different schemes that are realizable. In figure 2 we show all the possible schemes with resonant transitions involving a strong pump field and either one or two weak probe fields. As an illustration, we may consider the driven Λ -scheme (figure 2e) which includes as a particular case the driven h-scheme (figure 2b). The field equations (17) and (18)

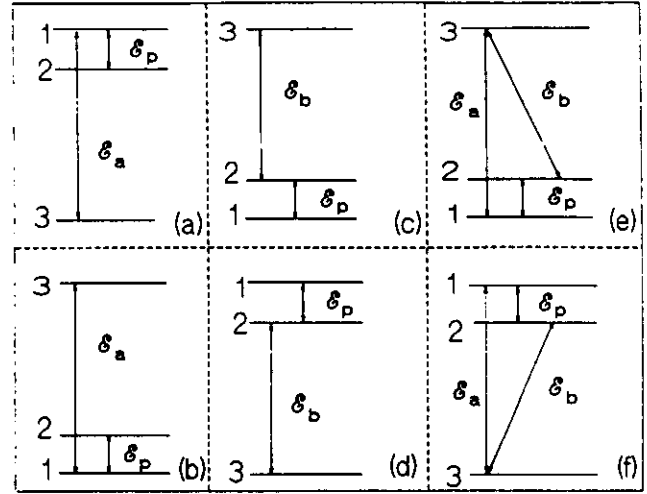


Figure 2. Six possible configurations of three-level media interacting with one strong pump field and one or two weak probe fields, all transitions being resonant.

are not modified. The matter equations become

$$\frac{\partial \sigma_{31}}{\partial t} = -\sigma_{31}[\gamma_{31} + i(\omega_3 - \omega_1 + \omega_a)] + i(xn_{13} + \beta\sigma_{21} - \gamma_p\sigma_{32}), \quad (29)$$

$$\frac{\partial \sigma_{32}}{\partial t} = -\sigma_{32}[\gamma_{32} + i(\omega_3 - \omega_2 + \omega_b)] + i(x\sigma_{12} + \beta n_{23} - \gamma_p^*\sigma_{31}), \quad (30)$$

$$\frac{\partial \sigma_{21}}{\partial t} = -\sigma_{21}[\gamma_{21} + i(\omega_2 - \omega_1 + \omega_a - \omega_b)] + i(\beta^*\sigma_{31} - x\sigma_{23} + \gamma_p n_{12}), \quad (31)$$

$$\begin{aligned} \frac{\partial \rho_{11}}{\partial t} &= R_1 + 2 \text{Im}(x\sigma_{13} + \gamma_p\sigma_{12}), \\ \frac{\partial \rho_{22}}{\partial t} &= R_2 - 2 \text{Im}(\beta\sigma_{23} - \gamma_p^*\sigma_{12}), \end{aligned} \quad (32)$$

where γ_p is proportional to the pump field amplitude ϵ_p . For the linear regime, the strong pump field can be treated as a constant. In a nonlinear theory, it will also be described by a propagation equation similar to (17). The linear stability analysis is carried out in the same way as for equations (17–22) and results in a gain condition that can be verified in two ways.

3.2.1. The first mechanism

If both transitions $1 \leftrightarrow 3$ and $2 \leftrightarrow 3$ are dipole allowed, there are two possible mechanisms through which AWI can occur. The first mechanism leads to the condition (24) for the low-frequency coherence. The difference,

however, is that in the present case all matrix elements of the density operator depend on the pump field, so that the condition that the low-frequency coherence σ_{21} be larger than some lower bound becomes a condition on the pump field amplitude. Explicitly, the scaled pump amplitude $x = |R\gamma_p/\gamma_{21}|$ must verify the inequalities $x_- < x < x_+$ where x_{\pm} are the real roots of

$$x^2 + x\left(N_{13} + N_{23} - \frac{N_{12}^2\gamma_{21}}{R}\right) + N_{13}N_{23} = 0, \quad (33)$$

where $N_{pq} = N_p - N_q$ and with the definition

$$R = 2 \frac{N_{13}T_1^{12}T_1^{23} + N_{23}T_1^{21}T_1^{13}}{N_1T_1^{23} + N_2T_1^{31} + N_3T_1^{12}}. \quad (34)$$

If we assume that $\gamma_{31} = \gamma_{32}$ it can be shown that $x_- < x_+$ by using the inequalities (15) and therefore there is a domain of AWI. Note that, if we use only the two-level inequality $2/T_2^{12} \geq 1/T_1^{12}$, we find $x_- > x_+$ and there is no possibility of AWI. This shows how critical the inequality (15) is. It also indicates that the domain in parameter space where AWI occurs is fairly small. Given the link established between the gain condition (24) and the trap and interacting states, it is easy to understand that, in the present scheme, the pump field excites into the system the amount of low-frequency coherence necessary to reach the gain threshold by bringing an adequate number of atoms in the trap state.

3.2.2. The second mechanism

However, if one of the two transitions $1 \leftrightarrow 3$ or $2 \leftrightarrow 3$ is forbidden, only the second mechanism of AWI is possible. Suppose that the allowed transition is $1 \leftrightarrow 3$. In that case, the second mechanism is described by the following sequence of elementary processes.

- (a) The low-frequency field is coupled to the population difference n_{12} to induce the low-frequency coherence σ_{12} .
- (b) The low-frequency coherence is coupled to the optical field α to induce the optical coherence σ_{23} at the forbidden transition.
- (c) The coherence σ_{23} is coupled with the pump field γ_p to induce the optical coherence σ_{31} .
- (d) The optical coherence σ_{31} is the source of the field x in the propagation equation.

Through this sequence of processes which combine anti-Stokes scattering and quantum interference, the pump field establishes a coherence between the two states 2 and 3 despite the absence of an allowed transition. The gain condition for this process is

$$|\gamma_p|^2 > \frac{N_{13}\gamma_{12}\gamma_{23}}{N_{12} - R\gamma_{23}}. \quad (35)$$

A necessary condition to achieve gain is that $N_{12} > R\gamma_{23}$ which involves the initial populations and four decay rates. It was shown by Kocharovskaya and Mandel (1991a) that the gain condition (35) can be fulfilled without population inversion. The main problem with this scheme is that the condition (35) does not correspond to a population inversion, neither in the dressed state basis nor in any other basis that could be found. The source of amplification in this case remains an open question.

It is clear from figure 2 that many other resonant schemes are possible. There are also degenerate schemes, non-resonant schemes and schemes that involve states in the continuum. This review is not the right place for an exhaustive description of all these alternative schemes nor can we cite all researchers that have contributed to the field by explicitly naming and commenting on their papers (especially since there is still much debate and acrimony among some colleagues on who did what and who did it first!). Therefore a long list of references is given at the end of this review, which will help the interested reader to enter the field.

Another restriction is to be noted. We have concentrated the discussion on AWI, neglecting the problems related to LWI. This is because there are very few papers that deal with the theory of lasers without inversion and no experimental support exists as of now. The first paper that treats in a consistent way the nonlinear equations predicting a steady-state LWI was by Kocharovskaya *et al.* (1990a). Agarwal (1991a) has calculated the linewidth of a LWI and showed that it may have a width narrower than that of conventional lasers. Finally, the recent paper by Gheri and Walls (1992) shows that a LWI may also be a source of sub-Poissonian statistics and squeezed light.

4. Experimental results

The first experimental report on AWI was by Gao *et al.* (1992). The experiment was performed using sodium atoms whose hyperfine components $F = 2$ and 1 of the ground state $^3S_{1/2}$ are coupled to the excited state $^3P_{1/2}$ by an intense coherent pump field at 589.6 nm. A weak tunable field probes the gain in the frequency domain near 589 nm. The status of this experiment is still fuzzy because the amplification reported by these authors is not accompanied by clear evidence of a lack of inversion. However, one should credit this experiment with stimulating further research.

The first clear-cut evidence of AWI has been reported by Nottelmann *et al.* (1993). These authors used a scheme based on Zeeman coherence as shown on figure 3. It is a generalization of the Λ -scheme with three lower levels

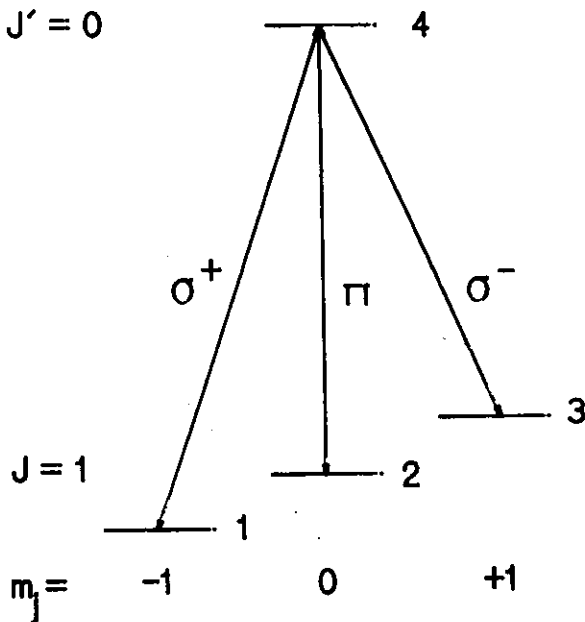


Figure 3. Samarium energy level and excitation schemes (after Nottelmann *et al.* 1993).

connected to one upper level. The atoms are subjected to a static magnetic field. The level energies are $E_1 = \hbar(\omega_2 - \Omega)$, $E_2 = \hbar\omega_2$, $E_3 = \hbar(\omega_2 + \Omega)$ and $E_4 = \hbar(\omega_2 + \omega_a)$ which defines the atomic frequency as $\omega_4 - \omega_2$ and the Larmor frequency as $\Omega = |\omega_1 - \omega_2| = g\mu_B B/\hbar$. The principle of the experiment is as follows. A low-frequency coherence ρ_{13} is created by means of a periodic train of picosecond pulses with r.f. period T_p which excites only the σ^+ and the σ^- transitions and therefore provides the source of interference. After each pulse, the low-frequency coherence ρ_{13} oscillates with the frequency 2Ω . For good control of the experimental situation, the magnetic field is chosen such that $\pi/\Omega = T_p/n$, $n = 1, 2$ or 4 . A test picosecond pulse, similar to those of the train, follows the last pulse of the train after a delay of $T_p/4$. Depending on the value of n , three different responses are possible.

- (1) If $n = 1$, the test pulse arrives when $\text{Re}(\rho_{13}) = 0$ and is attenuated since only $\text{Re}(\rho_{13})$ contributes to the energy transfer and there is no population inversion. In fact, there is never inversion in this experiment in the sense that the inequalities $\rho_{kk} < \rho_{44}$, $k = 1, 2, 3$ always hold.
- (2) If $n = 2$, $\text{Re}(\rho_{13})$ is positive and maximum, leading to an even stronger attenuation as for $n = 1$.
- (3) If $n = 4$, $\text{Re}(\rho_{13})$ is negative and minimum, leading to a maximum of amplification.

However, during the time interval $T_p/4$, the optical coherences ρ_{14} and ρ_{34} relax almost completely and the population in the upper level 4 also relaxes partially.

Since the gain is proportional to the upper-level population, it is useful to compensate for the transient decay of the upper level, although without creating an inversion. A populating picosecond pulse is sent into the system just before the test pulse arrives. The populating pulse has a polarization orthogonal to the other pulses such that it drives only the π transition without affecting the two σ transitions. Typically, the experiment is carried out with $T_p = 13.214$ ns and the populating pulse is sent 175 ps before the test pulse. Inversion at any time is prevented by carefully monitoring the area under each pulse. The experimental results are summarized in figure 4 which shows the intensity of the test pulse against time. In figure 4, each curve is the mean of 16 single curves and each single curve is the mean of 5500 test pulses. This procedure is followed to eliminate fluctuations.

The second experiment reporting AWI was performed by Fry *et al.* (1993) using the D_1 line of atomic sodium. The structure is quite complicated, as shown in figure 5 a. However, the basic principle of the experiment is easy to understand with the help of the generic diagram in figure 5 b. Here also, two strong fields are used to drive the $^3S_{1/2}(F=2) \rightarrow ^3P_{1/2}(F=2)$ and the $^3S_{1/2}(F=1) \rightarrow ^3P_{1/2}(F=2)$ transitions. They are the source of the coherence between the two lower levels b and b' and serve to bring atoms in the trap state $\Psi_{\text{trap}} \propto \phi_b - \phi_{b'}$ (assuming the two Rabi frequencies Ω_1 and Ω_2 are equal). This has been verified for this set-up by switching on and off one of the fields. As a result of the switching off at one field, the other field is attenuated because atoms in the trap state can again interact with the remaining field and absorb it since there is no population inversion. This is a transient evolution, until the new steady state is reached. Thus the interference process traps atoms in the state Ψ_{trap} only when the two fields are on. Then a weak populating field is applied to bring atoms in the upper state a of figure 5 b without inducing population inversion. As a result, a transient AWI of Ω_1 and Ω_2 occurs, before a redistribution of the atoms among the energy levels decreases the intensity through normal absorption. To confirm the role of interference, a similar experiment was performed in which the field Ω_2 is switched off as the populating field is applied. The net result is an attenuation of the field Ω_1 which confirms the essential role of the two fields to induce the atomic interference via the low-frequency coherence. Although the simple model of figure 5 b is a useful guide, a careful analysis and study of this experiment requires the consideration of the full diagram shown in figure 5 a. Note that here also the populating field is decoupled from the transitions excited by the strong fields by exploiting the field polarizations; the strong and the weak fields in this experiment have opposite polarizations.

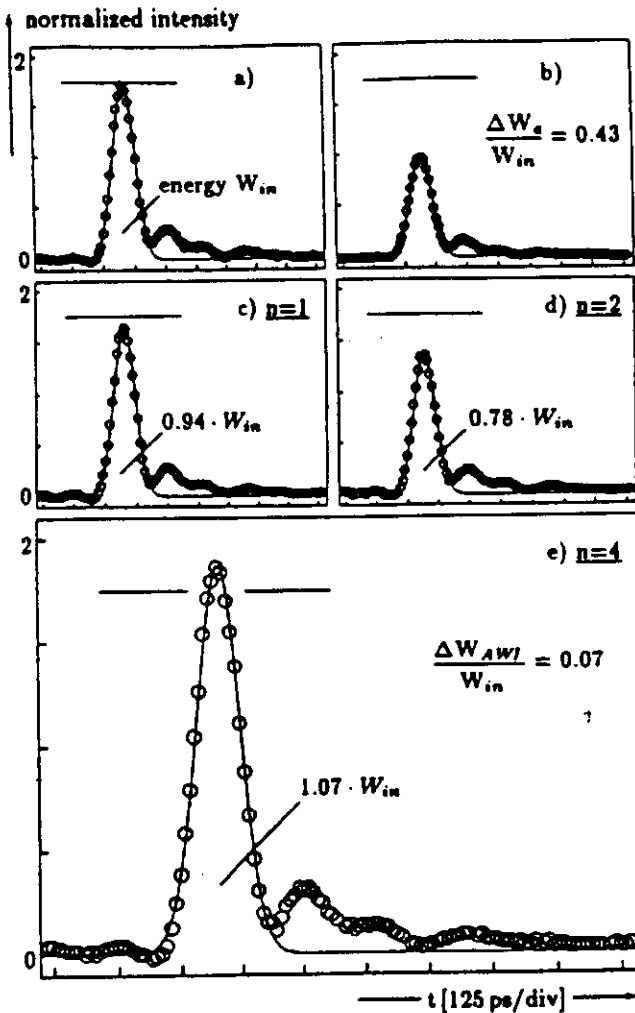


Figure 4. Experimental results obtained on samarium: intensity of the time-resolved test pulse (from Nottelmann, A., Peters, C., and Lange, W., 1993, *Phys. Rev. Lett.*, 70, 1783, with permission). (a) Magnetic field is chosen to induce electromagnetic transparency; (b) absorption of the test pulse in the absence of preparation pulses; (c) magnetic field is on with $\pi/\Omega = T_p$; weak absorption; (d) magnetic field is on with $\pi/\Omega = \frac{1}{2}T_p$; strong absorption; and (e) magnetic field is in with $\pi/\Omega = \frac{1}{4}T_p$; AWI.

The third piece of experimental evidence of AWI was proposed by van der Veer *et al.* (1993) using cadmium vapour in a magnetic field (figure 6). A linearly polarized pulse Π_1 excites the ground state $5s^2 \ ^1S_0$ to a coherent superposition of the states $5s5p \ ^3P_1$ with $m = \pm 1$ corresponding to energies E_{+1} and E_{-1} and wavefunctions $\phi_m = \pm 1$. In this experiment, the state P_1 with $m = 0$ does not play any role. The splitting is due to the magnetic field. Note that this time we have a V-scheme rather than the usual Λ -scheme. The two superposition states excited by the pulse Π_1 are $\phi_{\pm} \propto \phi_{m=1} \pm \phi_{m=-1}$. In this case, the interacting state is ϕ_+ while ϕ_- is the

non-interacting state. The two sublevels of the P_1 state are excited by the same broad-band laser pulse. An identical result would have been obtained using a resonant bichromatic field. What should be noticed here is that this state preparation realizes precisely the conditions that lead to the transition probability (5). Hence there is an oscillation between the two states ϕ_+ and ϕ_- at the low frequency $(E_{+1} - E_{-1})/\hbar$. At a time such that ϕ_+ is populated and ϕ_- is empty, a second light pulse Π_2 , whose polarization is perpendicular to that of Π_1 , brings some of the atoms of ϕ_+ into the upper state $5s6s \ ^3S_1$ and a weak probe beam couples the 3S_1 and the ϕ_- state. The difficulty in this experiment is the timing of the last two pulses with respect to the first pulse. In particular, the delay and the power of the second pulse must be controlled such that no population inversion is created between the 3S_1 and the 3P_1 states, while the probe pulse must reach the system when the atoms are in the ϕ_- state to prove AWI.

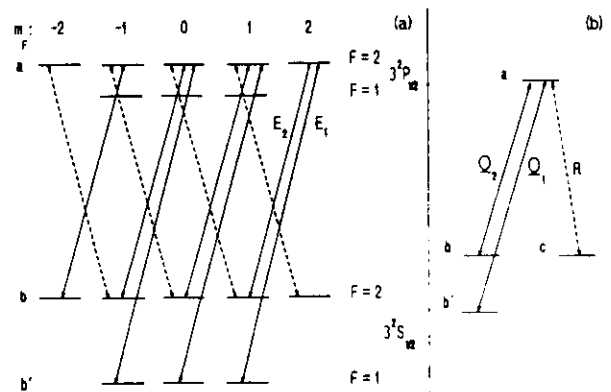


Figure 5. Sodium energy level and excitation schemes: (a) complete scheme; and (b) principle of the experiment (after Fry *et al.* 1993).

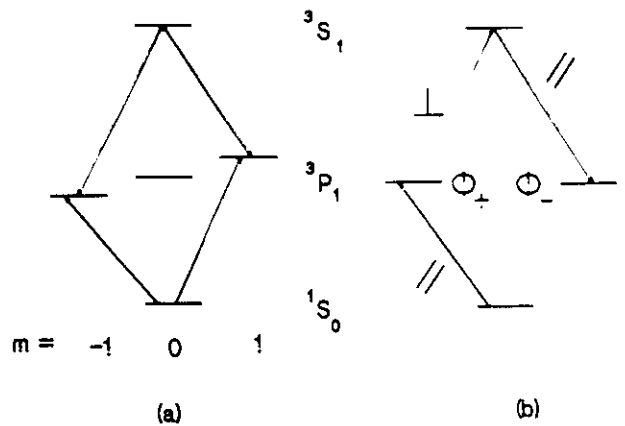


Figure 6. Energy level diagram of cadmium: (a) bare energy levels; and (b) superposition state energy levels (after van der Veer *et al.* 1993).

5. A useful concept?

A laser without population inversion but with a steady output intensity (Kocharovskaya *et al.* 1990) still remains an elusive goal. However, LWI and AWI belong to a young field of research in which coherent fields are used to manipulate atoms and to exploit interference of atomic wavefunctions. For instance, a recent set of experiments was reported by Hakuta *et al.* (1991), Boller *et al.* (1991) and Field *et al.* (1991) in which electromagnetically induced transparency was reported. In these experiments, gain is not achieved (and not sought) but a drastic reduction in the absorption due to coherent processes takes place as predicted by Kocharovskaya and Khanin (1986). As shown by Harris (1993) a method of matched pulses may lead to a strictly zero absorption in an otherwise optically thick medium. Still another effect was predicted by Scully (1991); a medium can be prepared in a state such that the frequencies at which the absorption vanish and the refractive index is maximum coincide. This could lead to a number of exotic applications such as a laser accelerator or the detection of ultra-weak magnetic fields.

In each of these examples, an external source is used to excite a suitable combination of atomic states in which the wave nature of matter can be used to create states with unexpected properties. The surprise is that *classical* fields can *spontaneously* create these combination states. Coherent optics is progressively paving the way to coherent atomics.

Acknowledgments

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