Propagation of Intense Coherent Light Waves in Resonant Media

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The propagation of a strong coherent "pump" wave traveling through a resonant medium is discussed. The medium is assumed to consist of identical stationary atoms, and the pump-wave frequency is assumed to coincide exactly with the resonance frequency for transitions between a particular pair of atomic states. A previously developed theory of the response of a strongly driven atom to a weak nearly resonant "signal" field is used to deduce the existence of a coupling between any two waves travelling parallel to the pump wave whose (nearly equal) frequencies sum to twice the pump-wave frequency. In the limit of very intense (highly saturating) pump fields, the coupling between the two waves of nearby frequencies leads to amplification of both waves if their frequencies lie within an interval about the pump-wave frequency equal to the Rabi frequency of population inversion. The basic process, which is roughly described by travelling-wave parametric-amplifier equations of motion, consists of the absorption of two pump-field photons followed by their emission at different nearby frequencies, and thus implies a frequency instability of the initially coherent pump wave.

The propagation of a light wave through a homogeneous medium leads to a number of interesting effects when the wave is intense enough and near enough in frequency to a particular atomic transition frequency to cause an appreciable modulation in the populations of the coupled atomic states. In many of the more commonly studied effects, e. g., in the cases of photon echo and of self-induced transparency, [1] the incident light is assumed to consist of a pulse of duration short compared to the atomic lifetime, so that decay processes are of minor importance during the brief interval within which the atom interacts with the incident field. In this paper we treat the opposite limit, that of infinite pulse duration, in which the incident field may be assumed, in lowest order, to oscillate harmonically with constant amplitude throughout the interval in question. In particular, we study the frequency stability of the process by assuming that in addition to the (strong) incident "pump" wave, a weak perturbing or "signal" wave of nearby frequency is initially present, and then evaluating the propagation characteristics of the signal wave.

The effect of a weak signal field on an atom driven by a strong pump field has been found. [2] When, as in the present case, both fields oscillate at frequencies near the resonance frequency for transitions between a particular pair of states, it was found that the signal field induces two components of comparable magnitude in the atomic dipole moment, one oscillating at the signal-field frequency ν, and the other at a frequency symmetrically displaced relative to the pump frequency ω, i. e., at the frequency $\omega-(\nu-\omega)$. The first component determines the average rate of absorption of energy from the signal field, which was found for certain values of the signal-field frequency to be represented by a negative number, corresponding to amplification rather than to absorption *per se*. The second, symmetrically placed frequency component in the induced dipole moment has no average effect, in general, in a medium consisting of many similar atoms, since the signal and pump phases are uncorrelated, in an average sense, throughout the medium.

An exception to this rule occurs, however, when the signal and pump fields travel in the same direction. [3] In that case, as we shall show, the signal field becomes parametrically coupled to a complementary field oscillating at the symmetrically placed frequency, and both fields become amplified because of their mutual interaction as well as the single-field amplification effect discussed previously. It is clear that the coupling between the two fields, which has the same general form as that which occurs in the parametric amplifier, must lead to an amplification of the zero-point or spontaneous-emission field of the pumped atoms. (This occurs within a frequency interval roughly comparable to the Rabi[4] frequency of population oscillation caused by the pump field.) Pairs of photons are in effect transferred from the pump field to two fields traveling parallel to the pump field, with frequencies which sum to twice the pump frequency. Our results thus demonstrate a frequency-instability in strong coherent light waves traveling in resonant media.

Let us consider an infinite homogeneous medium consisting of identical two-level atoms, each with a ground state $|0\rangle$ and a single excited state $|1\rangle$. We assume that a strong coherent "pump" wave is traveling through the medium in the z direction,

$$E_P(z,t)=\mathcal{E}_0\, e^{ikz-i\omega t}+\mathcal{E}_0^*\, e^{-ikz+i\omega t}, \qquad (1)$$

with electric polarization in the x direction. We take the field to be exactly on resonance, $\omega=(E_1-E_0)/\hbar$, and assume its propagation to be essentially unaffected by the medium, $k=\omega/c$.

A weak signal field

$$E(\vec{r},t)=\mathcal{E}(\vec{r})e^{-i\nu t}+\mathcal{E}^*(\vec{r})e^{i\nu t} \tag{2}$$

oscillating at a frequency $\nu=\omega+\Delta\nu$ close to the resonance frequency ω was shown in Ref. 2 to induce oscillating components[5]

$$\langle\mu(t)\rangle=\mu_+e^{-i(\omega+\Delta\nu)t}+\mu_-e^{-i(\omega-\Delta\nu)t}+\text{c. c.} \tag{3}$$

in the expectation value of the dipole moment operator for any atom in the medium. The induced volume density of electric polarization $P(\vec{r})$ may be expressed by means of generalized time- and position-dependent linear susceptibilities[6] $\chi_0(\nu)$ and $\chi_{-2}(\nu,z)e^{2i\omega t}$ as

$$P(\vec{r},t)=\chi_0(\nu)\mathcal{E}(\vec{r})e^{-i\nu t}+[\chi_{-2}(\nu,z)e^{2i\omega t}]^*\mathcal{E}^*(\vec{r})e^{i\nu t}+\text{c. c.}, \tag{4}$$

where the function $\chi_{-2}(\nu,z)$ has the form

$$\chi_{-2}(\nu,z)=\chi_{-2}(\nu)e^{-2ikz}. \tag{5}$$

The quantities $\chi_0(\nu)$ and $\chi_{-2}(\nu)$ are readily found from the analysis of Ref. 2 to be given by the relations

$$\chi_0(\nu)=(i/\hbar)N\mu_{01}\mu_{10}(\bar{n}_0-\bar{n}_1)\times\left(\frac{(-i\Delta\nu+\kappa)(-i\Delta\nu+\kappa')+\frac{1}{2}i\Omega^2\Delta\nu/\kappa'}{f(-i\Delta\nu)}\right), \tag{6a}$$

$$\chi_{-2}(\nu)=(2i/\hbar)N\mu_{10}\mu_{10}(\bar{n}_0-\bar{n}_1)\times\left(\frac{\mathcal{E}_0\cdot\mu_{10}}{\hbar}\right)^{*2}\frac{(-i\Delta\nu+2\kappa')}{\kappa' f(-i\Delta\nu)}, \tag{6b}$$

where N is the number density of atoms in the medium, μ_{10} is the dipole matrix element connecting the two atomic states $|0\rangle$ and $|1\rangle$, $\bar{n}_0$ and $\bar{n}_1$ are the corresponding equilibrium occupation numbers in the presence of the pump field, κ and κ' are the diagonal and off-diagonal relaxation rates, respectively, and $\Omega=2|\mathcal{E}_0\cdot\mu_{10}/\hbar|$. The function f is defined in terms of these parameters as

$$f(s)=(s+\kappa')[(s+\kappa)(s+\kappa')+\Omega^2]. \tag{7}$$

The susceptibilities evaluated in Eqs. (6) are assumed to be small compared to unity, and the function $\mathcal{E}(\vec{r})$ accordingly has the approximate spatial dependence $e^{i\vec{k}'\cdot\vec{r}}$, where $|\vec{k}'|=\nu/c$. It is clear then from Eq. (5) that the second term on the right-hand side of Eq. (4), which has the time dependence $e^{-i(\omega-\Delta\nu)t}$, will have the proper spatial dependence of a freely propagating plane electromagnetic wave only if $\vec{k}'$ points in the positive z direction. A complementary wave oscillating at the frequency $\omega-\Delta\nu$ is thus generated if the signal and pump fields propagate in the same direction, which is then the propagation direction for the generated wave as well.[3,3a]

Let us consider, then, only fields that propagate in the positive z direction, and take all electric polarizations to lie in the x direction. Such fields satisfy the equation (in rationalized units)

$$\left(\frac{\partial^2}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)E(z,t)=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}P(z,t), \tag{8}$$

which may be approximated for the case of nearly free propagation as

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)E(z,t)=-\frac{1}{2c}\frac{\partial}{\partial t}P(z,t). \tag{9}$$

The Fourier transform functions

$$\tilde{\mathcal{E}}(z,\nu)\equiv(2\pi)^{-1/2}\int_{-\infty}^{\infty}dt\,e^{i\nu t}E(z,t)\quad(\nu>0), \tag{10a}$$

$$\tilde{\mathcal{P}}(z,\nu)\equiv(2\pi)^{-1/2}\int_{-\infty}^{\infty}dt\,e^{i\nu t}P(z,t)\quad(\nu>0) \tag{10b}$$

then obey the equation

$$\left(\frac{\partial}{\partial z}-\frac{i\nu}{c}\right)\tilde{\mathcal{E}}(z,\nu)=\frac{i\nu}{2c}\tilde{\mathcal{P}}(z,\nu) \tag{11}$$

at every point in space. Let us separate out the free propagation of the field by introducing the definitions

$$\tilde{\mathcal{E}}(z,\nu)\equiv e^{i\nu z/c}\tilde{\mathcal{E}}'(z,\nu),\qquad \tilde{\mathcal{P}}(z,\nu)\equiv e^{i\nu z/c}\tilde{\mathcal{P}}'(z,\nu). \tag{12}$$

The primed functions then obey the equation

$$\frac{\partial}{\partial z}\tilde{\mathcal{E}}'(z,\nu)=\frac{i\nu}{2c}\tilde{\mathcal{P}}'(z,\nu)\approx\frac{i\omega}{2c}\tilde{\mathcal{P}}'(z,\nu), \tag{13}$$

the approximation holding for frequencies $\nu=\omega+\Delta\nu$ near the resonance frequency ω. The polarization function defined by Eq. (10b) in that case is given, according to Eq. (4), by the relation

$$\tilde{\mathcal{P}}(z,\omega+\Delta\nu)=\chi_0(\omega+\Delta\nu)\tilde{\mathcal{E}}(z,\omega+\Delta\nu)+\chi_{-2}^*(\omega-\Delta\nu,z)\tilde{\mathcal{E}}^*(z,\omega-\Delta\nu). \tag{14}$$

It follows from Eq. (5) and the relation $k=\omega/c$ that this relation, when expressed in terms of the primed functions defined by Eqs. (12), becomes

$$\tilde{\mathcal{P}}'(z,\omega+\Delta\nu)=\chi_0(\omega+\Delta\nu)\tilde{\mathcal{E}}'(z,\omega+\Delta\nu)+\chi_{-2}^*(\omega-\Delta\nu)\tilde{\mathcal{E}}'^*(z,\omega-\Delta\nu), \tag{15}$$

the z independence of the coefficients χ_0 and χ_{-2}^* reflecting the phase-matching condition associated with parallel propagation.

By substituting Eq. (15) into Eq. (13) and evaluating the resulting equation at the two frequencies

$$\nu_+\equiv\omega+\Delta\nu\quad\text{and}\quad\nu_-\equiv\omega-\Delta\nu, \tag{16}$$

we find the relations

$$\frac{\partial}{\partial z}\,\tilde{\mathcal{E}}'(z,\nu_+)=a(\Delta\nu)\,\tilde{\mathcal{E}}'(z,\nu_+)+b(\Delta\nu)\,\tilde{\mathcal{E}}'^*(z,\nu_-)\,,$$
$$\frac{\partial}{\partial z}\,\tilde{\mathcal{E}}'(z,\nu_-)=a^*(\Delta\nu)\,\tilde{\mathcal{E}}'(z,\nu_-)+b^*(\Delta\nu)\,\tilde{\mathcal{E}}'^*(z,\nu_+)\,, \tag{17}$$

where the coefficients $a(\Delta\nu)$ and $b(\Delta\nu)$ are defined as

$$a(\Delta\nu)=\tfrac{1}{2}ik\,\chi_0(\nu_+)=-\tfrac{1}{2}ik\,\chi_0^*(\nu_-)\,,$$
$$b(\Delta\nu)=-\tfrac{1}{2}ik\,\chi_{-2}(\nu_+)=\tfrac{1}{2}ik\,\chi_{-2}^*(\nu_-)\,. \tag{18}$$

The coupled equations (17) for the two functions $\tilde{\mathcal{E}}'(z,\nu_+)$ and $\tilde{\mathcal{E}}'(z,\nu_-)$ may be roughly described as representing traveling-wave parametric amplification. The solutions to Eqs. (17) and (12) are

$$\tilde{\mathcal{E}}(z,\nu_+)=e^{(a+i\nu_+/c)z}\times[\tilde{\mathcal{E}}(0,\nu_+)\cosh bz+\tilde{\mathcal{E}}^*(0,\nu_-)\sinh bz]\,,$$
$$\tilde{\mathcal{E}}(z,\nu_-)=e^{(a^*+i\nu_-/c)z}\times[\tilde{\mathcal{E}}(0,\nu_-)\cosh b^*z+\tilde{\mathcal{E}}^*(0,\nu_+)\sinh b^*z]\,. \tag{19}$$

It is clear from these relations that the waves oscillating at the two frequencies ν_+ and ν_- will become amplified as they travel in the positive z direction if either of the two quantities

$$\zeta_+(\Delta\nu)\equiv\mathrm{Re}[a(\Delta\nu)+b(\Delta\nu)] \tag{20a}$$

or

$$\zeta_-(\Delta\nu)\equiv\mathrm{Re}[a(\Delta\nu)-b(\Delta\nu)] \tag{20b}$$

is positive. We find from Eqs. (18), (6), and (7) that the real part of a (which is proportional to the negative of the absorption function evaluated in Ref. 2) is given in the limit of strong pump fields by the relation

$$\mathrm{Re}\,a(\Delta\nu)=\frac{\tfrac{1}{4}Nk|\mu_{10}|^2(\bar{n}_0-\bar{n}_1)\Omega^2}{\hbar\kappa'}\times\left(\frac{-(\Delta\nu)^4+\Omega^2(\Delta\nu)^2-2\kappa\kappa'^3}{|f(-i\Delta\nu)|^2}\right)\quad(\Omega\gg\kappa,\kappa')\,, \tag{21a}$$

while the real part of b is given in the same limit by the relation

$$\mathrm{Re}\,b(\Delta\nu)=\frac{\tfrac{1}{4}Nk|\mu_{10}|^2(\bar{n}_0-\bar{n}_1)\Omega^2}{\hbar\kappa'}\times\left(\frac{-(\Delta\nu)^4+\Omega^2(\Delta\nu)^2+2\Omega^2\kappa'^2}{|f(-i\Delta\nu)|^2}\right)\quad(\Omega\gg\kappa,\kappa')\,. \tag{21b}$$

It follows directly from these relations that of the two parameters defined by Eqs. (20), ζ_- is negative for all values of $\Delta\nu$, representing attenuation rather than amplification, while ζ_+ is found with the aid of Eq. (7) to be well approximated by the relation

$$\zeta_+(\Delta\nu)=\frac{\tfrac{1}{2}Nk|\mu_{10}|^2(\bar{n}_0-\bar{n}_1)\Omega^2}{\hbar\kappa'}\times\left(\frac{\Omega^2-(\Delta\nu)^2}{[(\Delta\nu-\Omega)^2+\bar{\kappa}^2][(\Delta\nu+\Omega)^2+\bar{\kappa}^2]}\right)\quad(\Omega\gg\kappa,\kappa'), \tag{22}$$

where $\bar{\kappa}=\tfrac{1}{2}(\kappa+\kappa')$. The parameter ζ_+ is thus positive for values of $\Delta\nu$ lying within the interval $-\Omega<\Delta\nu<\Omega$, and the two components of the field oscillating at the frequencies $\omega+\Delta\nu$ and $\omega-\Delta\nu$ are consequently amplified in this frequency interval by the coupling between them which is produced by the pump field.

It is clear that this result implies that the initially coherent pump field is unstable, acquiring as it travels through the resonant medium frequency components of ever increasing magnitude within an interval equal to the Rabi frequency Ω about the central frequency ω. An initial excitation within this frequency interval is certainly present in the form of the spontaneous-emission field of the strongly pumped atoms.[7] The portion of this initial fluctuating field that is traveling in the same direction as the pump field will become amplified, in much the same way as vacuum fluctuations are amplified in the parametric amplifier.[8] An explicitly quantum-mechanical analysis of the spontaneous-emission process (which we do not present here) confirms the above interpretation of our semiclassical analysis, showing the inelastic components of the field radiated by the coherently pumped atoms to be due, in lowest order, to pairs of photons whose frequencies sum to twice the frequency of a pump-field photon, and whose propagation directions are strongly peaked in the direction of the pump wave.

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[1]See, for example, L. Matulic and J. H. Eberly, Phys. Rev. A **6**, 822 (1972), and references cited therein.

[2]B. R. Mollow, Phys. Rev. A **5**, 2217 (1972).

[3]The signal and pump fields travel in opposite directions in the technique of *laser-saturated absorption*. See, for example, M. D. Levenson and A. L. Schawlow, Phys. Rev. A **6**, 10 (1972). Theoretical analyses of this phenomenon are given by E. V. Baklanov and V. P. Chebotaev {Zh. Eksp. Teor. Fiz. **60**, 552 (1971) [Sov. Phys.-JETP **33**, 300 (1971)]} and by S. Harache and F. Hartmann, Phys. Rev. A **6**, 1280 (1972). No complementary field is generated in this case, and the basic result for the absorption of the signal field (before Doppler averaging) is directly derivable from the single-field absorption rate evaluated in Ref. 2.

[3](a) If the signal wave travels at a small angle relative to the pump wave, then the generated wave will travel in the complementary direction, thus conserving momentum as well as energy. The basic process considered here is in a sense the continuous-wave analog of the (short-pulse-limit) photon echo effect. It has been called "light-by-light scattering," and has been observed by R. L. Carman, R. Y. Chiao, and P. L. Kelley [Phys. Rev. Letters **17**, 1281 (1966)] in the case in which the nonlinear coupling is due to the molecular-orientation Kerr effect.

[4]I. I. Rabi, Phys. Rev. **51**, 652 (1937).

[5]Equations (3)–(6) of this paper follow directly from Eqs. (3.5a)

and (3.11) of Ref. 2, when the pump-field amplitude in the latter relations is multiplied by the phase factor e^{ikz}.

[6]A general treatment of time-dependent linear susceptibilities for systems driven near resonance will be presented by the author in a subsequent publication.

[7]B. R. Mollow, Phys. Rev. **188**, 1969 (1969); Phys. Rev. A **2**, 76 (1970). The analysis of these papers is based on a quantum regression theorem derived by M. Lax [Phys. Rev. **172**, 350 (1968), and references cited therein].

[8]A realistic analysis of the frequency instability we have described would have to take into account the nonzero angular width of the pump wave.

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Low-Temperature Specific Heats of Adsorbed Helium Monolayers in the Mobile Limit*

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Jackson's model for adsorbed helium monolayers is suitable for studying thermodynamic properties in the mobile limit. In particular, low-temperature specific heats can be calculated over a range of areal densities. The dominant contributions come from longitudinal surface phonons. We report here (i) an extension of Jackson's model to account for surface-phonon–surface-phonon interactions, (ii) computations of low-temperature specific heats to several coverages, and (iii) a comparison of the computed results to experiment.

I. ADSORBED HELIUM MONOLAYERS IN MOBILE LIMIT

Consider N helium atoms physically adsorbed on a smooth surface of a crystalline substrate. Let the adsorbing area be denoted by A. The system is then characterized by a single physical parameter: $n=N/A$, the areal density of the adsorbed layer.

Following our recent work published in Refs. 1 and 2, we make the preliminary approximation that the substrate serves no other purpose than to provide a static "external" field $V(\vec{r})$ to each adatom. The Hamiltonian of the system is then given by

$$H=\sum_{i=1}^{N}\frac{-\hbar^2}{2m}\nabla_i{}^2+\sum_{i=1}^{N}V(\vec{r}_i)+\sum_{1\leq i<j\leq N}v(r_{ij}), \qquad (1)$$

where $\vec{r}$ is a three-dimensional vector and $v(r)$ represents the adatom-adatom interaction potential. In this work the pairwise, central Lennard-Jones 6-12 potential is used.

To solve the Schrödinger equation

$$H\psi(1, 2, \ldots, N)=E\psi(1, 2, \ldots, N) \qquad (2)$$

for the ground state and the low-lying excited states, we proposed in Ref. 1 the employment of a set of correlated basis functions:

$$\phi_{\mu_1\vec{\nu}_1,\mu_2\vec{\nu}_2,\ldots,\mu_N\vec{\nu}_N}(z_1\vec{\rho}_1, z_2\vec{\rho}_2, \ldots, z_N\vec{\rho}_N) = F(1, 2, \ldots, N)P\left\{\prod_{i=1}^{N}\varphi_{\mu_i\vec{\nu}_i}(z_i\,\vec{\rho}_i)\right\}, \qquad (3)$$

where F denotes a symmetrical correlating factor which accounts for the short-ranged adatom-adatom correlations and $\varphi_{\mu\nu}(z\vec{\rho})$ stands for the wave functions describing the motion of a single adatom, i.e., the eigenfunctions of the single-particle Hamiltonian:

$$h(\vec{r})=\frac{-\hbar^2}{2m}\nabla^2+V(\vec{r}). \qquad (4)$$

The quantum number μ characterizes the bound states normal to the adsorbing surface (the z direction), and the two-component vector $\vec{\nu}$ characterizes motion parallel to that surface. For a realistic crystalline substrate, φ must clearly contain Bloch functions possessing the periodicity of the surface.

The subspace spanned by a set of basis functions of the type (3) with just one elementary excitation is of most importance when one considers low-temperature properties of the system. We take for this subspace $\mu_1=\mu$ and $\vec{\nu}_1=\vec{\nu}$, and $\mu_i=0$ and $\vec{\nu}_i=0$ for all $i\neq1$. The basis functions of interest are then given by

$$\phi_{\mu\vec{\nu},00,\ldots,00}=\sum_{i=1}^{N}\left[\frac{\varphi_{\mu\vec{\nu}}(z_i\vec{\rho}_i)}{\varphi_{00}(z_i\,\vec{\rho}_i)}\right]\phi_0, \qquad (5)$$

where

$$\phi_0\equiv\phi_{00,00,\ldots,00}=\prod_{i=1}^{N}\varphi_{00}(z_i\,\vec{\rho}_i), \qquad (6)$$

and $\vec{\rho}$ is a two-component vector in a plane parallel to the adsorbing surface.

In the special case of a completely uniform and homogeneous substrate, the single-particle functions become

$$\varphi_{\mu\vec{\nu}}(z\vec{\rho})=\chi_\mu(z)e^{i\vec{\nu}\cdot\vec{\rho}}, \qquad (7)$$

where $\vec{\nu}$ now represents a two-dimensional wave vector. Consequently Eq. (5) reduces to

$$\phi_{\mu\vec{\nu},00,\ldots,00}=\sum_{i=1}^{N}\left[\frac{\chi_\mu(z_i)e^{i\vec{\nu}\cdot\vec{\rho}_i}}{\chi_0(z_i)}\right]\phi_0, \qquad (8)$$