Propagating, Optical-Pumping Wave Fronts

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We have discovered a novel, spatially propagating wave front which is generated by laser pumping of an optically thick medium to a state of complete transparency. The wave front consists of a moving band of fluorescent light which marks the location of atoms which are being pumped. The wave-front velocity v, the photon flux I_0, the atomic number density N, and the number n of photons absorbed per atom are related by $v = I_0 \times (Nn)^{-1}$.

In this paper we shall describe the properties of a novel propagating wave front which can be generated in any optically thick medium that can be optically pumped to a state of complete transparency. Preliminary experimental studies of this new wave front have been carried out with laser-pumped sodium vapor.

The essence of the phenomenon is illustrated in Fig. 1. Spin-$\frac{1}{2}$ atoms with energy levels indicated in Fig. 1(c) can be pumped by circularly polarized light until complete spin polarization is achieved. As shown in Fig. 1(a), two circularly polarized laser beams, an intense right-circularly polarized beam R and a weaker left-circularly polarized beam L, are incident on a cell which contains so many atoms that it is optically thick. The laser beams are absorbed in a thin boundary layer of width α^{-1}, where α is the attenuation constant of the unpolarized atomic vapor. Negligible excitation occurs to the left of the boundary where the atoms are completely spin polarized and therefore transparent, and little excitation occurs to the right of the boundary layer because the laser light cannot penetrate much beyond the boundary. As the atoms in the boundary become optically pumped the boundary moves to the right. Clearly the boundary for the more intense beam will move faster because the atoms are pumped faster. Furthermore, the intense central portion of the beams will propagate more rapidly than the weaker outer part, and a convex, cone-shaped boundary will develop as the beams penetrate deeper into the vapor.

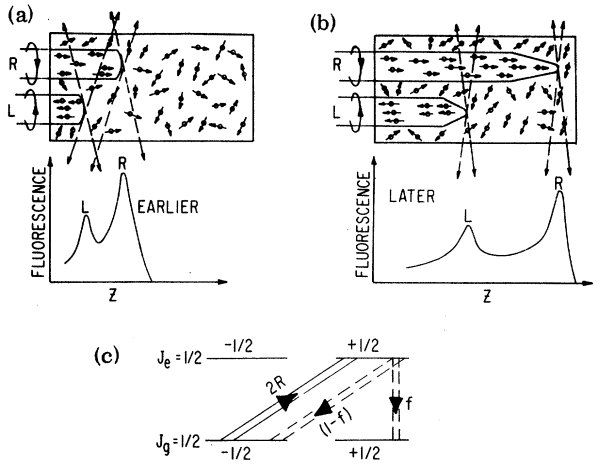

FIG. 1. Simple model for propagating, optical-pumping wave fronts.

A more quantitative understanding of the wave front may be gained from the simple model of Fig. 1. A combination of collisional effects and radiative decay causes a fraction f of spin-up excited atoms to decay to the spin-up sublevel of the ground state. The rate equations for the number densities of ground-state atoms are

$$\partial N(\downarrow)/\partial t = -2RN(\downarrow) + (1-f)2RN(\downarrow) = -2RfN(\downarrow), \quad (1)$$

$$\partial N(\uparrow)/\partial t = 2RfN(\downarrow). \quad (2)$$

The mean pumping rate R is related to the local photon flux I and to the mean optical absorption cross section σ by $R = I\sigma$. The rate R is assumed to be much smaller than the decay rate of the excited state, which therefore has negligible population. The attenuation of the pumping light is given by

$$\partial I/\partial z = -2N(\downarrow)\sigma I. \quad (3)$$

The boundary conditions on (1)–(3) are $I(z=0) = I_0$, the incident light intensity; and the atoms are initially unpolarized, i.e. $N(\downarrow, t=0) = N/2$, where $N = N(\downarrow) + N(\uparrow)$ is the total atomix number density. One can easily verify that the solution to (1)–(3), subject to these boundary conditions, is

$$I/I_0 = e^{\theta}[e^{\xi} + e^{\theta} - 1]^{-1}, \quad (4)$$

$$2N(\downarrow)/N = e^{\xi}[e^{\xi} + e^{\theta} - 1]^{-1}, \quad (5)$$

where $\theta = 2fR_0 t$, $\zeta = \alpha z$, and $\alpha = N\sigma$.

Note that we may integrate the local photon absorption rate, $2RN(\downarrow)$, to get the total number n of absorbed photons per atom as

$$n = \frac{1}{N}\int_0^\infty 2RN(\downarrow)\,dt$$

$$= \frac{-1}{fN}\int_0^\infty \frac{\partial N(\downarrow)}{\partial t}\,dt = \frac{1}{2f}. \quad (6)$$

We have used (1) and the boundary conditions on $N(\downarrow)$ to evaluate (6).

For $z \gg \alpha^{-1}$ and $t \gg (2fR_0)^{-1}$, Eqs. (4) and (5) imply that the local photon absorption rate is

$$2RN(\downarrow) \simeq \tfrac{1}{4}R_0 \operatorname{sech}^2[\tfrac{1}{2}\alpha(z - vt)], \quad (7)$$

where the propagation velocity is

$$v = I_0(Nn)^{-1}. \quad (8)$$

We have worked out the theory of optical-pumping wave fronts for more realistic models. Hyperfine structure and excited-state collisional effects lead to more complicated formulas than Eq. (7) for the pulse shape and Eq. (6) for n. However, Eq. (8) for the velocity remains valid and no qualitative changes occur in the theoretical prediction (7) that a wave front propagates with constant velocity v and without distortion through the vapor. Ground-state relaxation leads to the theoretical prediction of a dimly glowing wake of relaxing atoms behind the propagating wave front, a gradual attenuation and slowing down of the wave front, and a finite range of the front.

We have made a preliminary study of the propagating wave fronts with the apparatus shown in Fig. 2. A cylindrical glass cell, 70 mm long and 12 mm in diameter, is filled with a small amount of sodium metal, 10 Torr of N_2, and 600 Torr of He at 20°C. The cell is situated in a longitudinal magnetic field H_0 of about 10 G. An argon-ion–pumped tunable dye laser with intracavity etalon (Spectra Physics, Model 580) is the source of a linearly polarized laser beam tuned to the center of the D_1 absorption lines of sodium vapor at 5896 Å. The beam is split with a Wollaston prism and converted to left- (L) and right- (R) circularly polarized beams by a quarter-wave plate. The two beams have an angular divergence of 1° and a spatial separation of several millimeters in the cell. By rotating the prism the relative intensities of the two beams can be adjusted to any desired value. The cell is heated to about 220–270°C [$N \simeq (1\text{–}11)\times 10^{13}/\text{cm}^3$]. At these temperatures the vapor is optically thick and α^{-1} is only a few millimeters. However, a circularly polarized laser beam easily penetrates the entire 70-mm length of the cell because of the efficient pumping with D_1 light.[1] A lens is used to project an image of the cell with unit magnification onto the face of a photomultiplier tube. A narrow slit in the focal plane is used to isolate the fluorescence from a small region of the cell. The entire cell can be scanned by moving the slit and photodetector parallel to the cell axis.

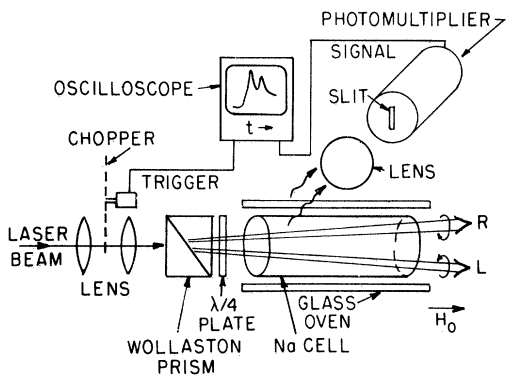

FIG. 2. Diagram of apparatus.

The 60-mW incident laser beam is suddenly turned on by the blade of a chopper wheel rotating at 13 Hz, and the sweep circuit of an oscilloscope is triggered on at the same time. The oscilloscope is used to display the detected fluorescent light intensity as a function of time after the laser beam is turned on. Typical data are shown in Fig. 3. In Fig. 3(a) the time evolution of the fluorescent intensity at the input face ($z = 0$) of the cell is shown. The fluorescence begins immediately and it rapidly decays as the atoms become spin polarized. Qualitatively similar transients are obtained with beam L or R separately or both together for $z = 0$. In Fig. 3(b) the fluorescence recorded at a distance $z = 2$ cm from the input face is shown. Only the beam L has been allowed to enter the cell and a time delay of 0.8 msec occurs before the beam reaches $z = 2$ cm and is visible at the photomultiplier tube. In Fig. 3(c) the fluorescence recorded at a distance $z = 4.65$ cm from the input face is shown. Both beams L and R are incident on the cell simultaneously but the strong beam R reaches the location $z = 4.65$ cm in less than half the time required for the slowly propagating weak beam L. In Fig. 3(d) the same experimental conditions prevail as in Fig. 3(c) except that the intensities of both beams

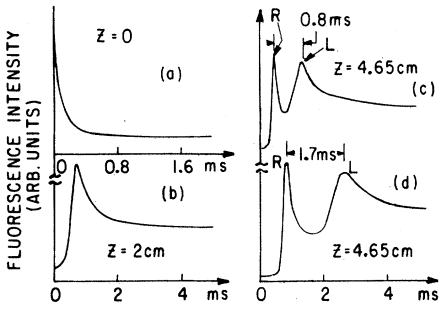

FIG. 3. Experimentally measured fluorescence for arrangement of Fig. 2. Power of the laser beam at the input face $z = 0$ is typically 40 mW. (a) Beam L alone at input face $z = 0$. (b) Beam L alone at $z = 2$ cm from input face. (c) Both L and R beams of different initial intensities launched simultaneously and observed at $z = 4.65$ cm from input face. (d) Same conditions as in (c) except both L and R are attenuated by 50% before entering the cell. The vertical scales in (a)–(d) are not the same.

have been attenuated by a factor of 2 with a neutral density filter. Note that both beams have been slowed down by a factor of 2 and the delay between the beams has been doubled, in agreement with the predictions of Eq. (8). Note that the propagation velocity of beam R of Fig. 3(c) is $v \simeq (4.65\ \text{cm})/(0.5\ \text{msec}) \simeq 9300$ cm/sec.

These newly discovered wave fronts have curious parallels to the pulses in self-induced transparency;[2] for example, the $\text{sech}^2[\frac{1}{2}\alpha(z - vt)]$ ideal pulse shape and the existence of a well-defined pulse area n from (6). However, the optical-pumping wave fronts are driven by a cw laser rather than by a pulsed laser as in self-induced transparency. Also optical-pumping transients could, in principle, be generated by incoherent light.

The optical-pumping wave fronts first drew our attention as unexpected anomalies in spin relaxation measurements. Consequently it is important to be aware of the possible existence of these wave fronts in spin-relaxation studies. The easily measured velocity v affords a direct way to determine the important and otherwise difficult-to-measure parameter n by means of (8). Finally, we note that similar optical-pumping transients should exist in many other systems, solid, liquid, or gaseous. Some of the saturable absorbers[3] used in Q switching or mode locking of lasers or in studies of optical bistability[4] should support propagating optical-pumping transients. The photodissociation waves predicted by Khartsiev[5] are closely analogous to the optical-pumping waves observed in these experiments. Indirect evidence for photodissociation waves has been reported by Powell and Ewing.[6]

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