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GIANT FARADAY ROTATIONS IN SELF-INDUCED TRANSPARENCY

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Giant Faraday and inverse Faraday effects accompanied by no energy loss are predicted for systems undergoing self-induced transparency. The size of these effects is calculated. These effects, and also the large velocity reduction which accompanies transparency, are shown to be intimately related to large energy storage in the resonant material system.

Macroscopic dipoles oscillating at optical frequencies have been observed by photon echoes¹ and, more recently, by self-induced transparency.^{2,3} This Letter points out that very large Faraday rotations and other unusual effects should be associated with the coherent excitation of these dipoles. For the investigation of the transparency phenomenon the Faraday effect provides a sensitive tool which will usefully supplement the transmission and delay measurements performed so far.

Self-induced transparency is characterized by continuous absorption and re-emission of radiation by a resonant material system in such a manner that steady-state optical pulses propagate. These so-called 2π pulses exhibit a signal velocity V which can be very much smaller than their phase velocity c/n. Giant Faraday rotation of a linearly polarized 2π pulse in an axial magnetic field is predicted to accompany this velocity reduction.

Consider the case of a Kramers doublet which splits in the magnetic field as shown in Fig. 1(a). The cross relaxation between the Zeeman sublevels is neglected since the duration τ of the optical pulse is shorter than the homogeneous part of the transverse relaxation time of the unsplit line. We are dealing therefore with a superposition of two noninteracting two-level systems.⁴ Moreover, it will be assumed that $kT \gg \hbar \omega_z$,

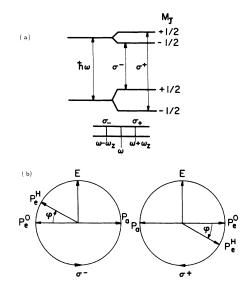


FIG. 1. (a) The resonant two-level system without and with an applied H field. The corresponding spectrum is shown schematically below the level diagram. (b) The field and polarization vectors in their rotating frames. The direction of rotation of the frame is indicated by an arrow at the lower part of the circles. where ω_z is the Zeeman shift [Fig. 1(a)], so that the ground-state Zeeman sublevels are equally populated, and N will denote the density of centers in one of the sublevels. In these conditions the Verdet constant ρ can be obtained from an extension of the theory of McCall and Hahn.² We allow for inhomogeneous broadening, which is described by the normalized spectral distribution function of the line $g(\Delta \omega)$ in the absence of magnetic field, where $\Delta \omega$ is measured from the angular frequency ω of the optical carrier. The linearly polarized pulse is decomposed into its left and right circularly polarized components having propagation vectors k_{\perp} and k_{\perp} , respectively. These are obtained from Eq. (11) of Ref. 2 where in the present case displaced distribution functions $g(\Delta \omega \pm \omega_z)$ are used. Using the expression $\rho = (k_+ - k_-)/2H$, where *H* is the applied magnetic field, one obtains

$$\rho = \frac{g\beta}{\hbar} \frac{4\pi N\omega p^2 \tau^2}{nc\hbar} \int_{-\infty}^{+\infty} \frac{1 - (\Delta\omega \tau)^2}{[1 + (\Delta\omega \tau)^2]^2} g(\Delta\omega) d(\Delta\omega),$$

where $\omega_Z/H=g\beta/\hbar=\gamma$ is the magnetogyric ratio, p is the x or y component of the transition dipole moment, and n is the refractive index of the host excluding the contribution from the two-level systems of interest. Defining the constant A $= 4\pi N\omega p^2/nc\hbar$, we see that in the case of a resonant line much narrower than $1/\tau$, we have ρ $= \gamma A \tau^2$. On the other hand, for a line with Gaussian profile $g(\Delta \omega) = T(2\pi)^{-1/2} \exp(-\frac{1}{2}T^2\Delta \omega^2)$, in the limit of large linewidth $1/T \gg 1/\tau$, we find ρ $= \gamma A T^2$.

A feeling for the size of the effect can be obtained by estimating the constant γA for the ${}^{2}S_{1/2}$ $\rightarrow {}^{2}P_{1/2}{}^{\circ}$ transition in potassium vapor at 300°C. With $N = 1.2 \times 10^{13}$ cm⁻³, $p = 7.2 \times 10^{-18}$ esu, $g = \frac{1}{2}(2 + \frac{2}{3}) = \frac{4}{3}$, and $\omega/2\pi c = 12\,988.7$ cm⁻¹, we obtain $\gamma A = 7.3 \times 10^{18}$ rad/Oe cm sec². For this system the Gaussian line profile is determined by Doppler broadening, and $T = 3.5 \times 10^{-10}$ sec. In the large-linewidth limit one obtains $\rho = \gamma AT^{2}$ = 0.9 rad/Oe cm, which is indeed enormous.

Similarly, the inverse Faraday effect⁵ is very large. A circularly polarized 2π pulse applied to the system of Fig. 1(a) generates an axial magnetization M which has the time dependence of the pulse intensity I. With $I = (nc/4\pi)\mathcal{E}^2$, where \mathcal{E} is the electric-field amplitude of the optical pulse, we find

$$\frac{M}{I} = \frac{\gamma A \tau^2}{\omega} \int_{-\infty}^{+\infty} \frac{g(\Delta \omega) d(\Delta \omega)}{1 + (\Delta \omega \tau)^2}.$$
 (2)

For a resonant line much narrower than 1/ au this

gives $M/I = \gamma A \tau^2/\omega$. In the other extreme of a Gaussian line with large linewidth, we find $M/I = (\frac{1}{2}\pi)^{1/2}\gamma A\tau T/\omega$. The inverse Faraday effect in a nonresonant system is characterized by the relation $M/I = \rho/\omega$.⁵ It is remarkable that this relation is preserved in the case of exact resonance for a 2π pulse of duration much shorter than the inverse linewidth of the medium.

In fact, under these conditions the magnitude of all these anomalous effects, including the velocity reduction, can be derived from simple physical arguments. The velocity reduction can be understood in terms of energy storage in the resonant centers. The physical picture is one where we consider separately the host medium and the resonant dipoles which act as source terms for the field. Any electromagnetic wave propagates in the host medium with a phase velocity c/n, where n is a characteristic of the host. If we denote by U_{em} the energy density in the electromagnetic wave, and by U_c the energy density accumulated in the excitation of the resonant centers, the total energy per unit cross-sectional area of a 2π pulse can be calculated in two ways. First, we integrate the average energy densities over the length $V\tau$ of the pulse and obtain $V\tau(U_{em}+U_c)$. Second, we integrate over time the energy flowing through a unit cross section. The flow of energy through this section occurs only through the electromagnetic wave and it takes place with a velocity equal to the phase velocity of the wave in the host medium. Therefore we obtain an energy $(c/n)\tau U_{em}$. By equating the two expressions we have

$$\frac{1}{V} = \frac{n}{c} + \frac{n}{c} \frac{U_c}{U_{\text{em}}}.$$
(3)

It is remarkable that this is, in another form, the exact expression derived by McCall and Hahn.^{2, 6} Clearly the proper averages to be used for the energy densities can only be calculated from the known pulse shape; however, since the time dependence of \mathcal{E}^2 and of the population inversion are identical,² the ratio $U_c/U_{\rm em}$ can be obtained anywhere in the pulse, for example from the peak values of the individual quantities. For exact resonance and infinite transverse relaxation time one has $(U_c)_{\rm peak} = N\hbar\omega$. With $(U_{\rm em})_{\rm peak} = n^2\hbar^2/4\pi p^2\tau^2$ we obtain from (3) $1/V \equiv n/c + A\tau^2$, which is identical to formula (10) of Ref. 2 under the same assumptions.

Similarly, the value of the Verdet constant can be estimated straightforwardly. The left and

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right circularly polarized components of the linearly polarized 2π pulse are shown in their respective rotating frames in Fig. 1(b). In the same figure the transverse components of the macroscopic electric dipole at coordinate z, in a layer of infinitesimal thickness dz, are shown in their positions of maximum absorption (subscript a) and maximum emission (subscript e), without and with magnetic field (superscript 0 and H, respectively). We use here the familiar pictorial representation.^{7,1} The z component of the effective dipole moment represents the population difference between upper and lower states; this component would be perpendicular to the paper and is not shown in Fig. 1(b). In the presence of an axial magnetic field, P rotates slightly faster than E for the σ^+ transition, and slightly slower than E for the σ^- transition, the difference in angular frequency being ω_z . This results in P_e^H being rotated away from P_e^0 by the angle $\varphi \sim 2\omega_z \tau$. Consequently, the field radiated by the dipole layer thickness dz is also rotated by φ . The total electromagnetic field at coordinate z + dz is the sum of that part of the field which was not absorbed by the dipoles carrying an energy which is $(c/n)U_{em}\tau$ minus an infinitesimal part that can be neglected and of that part of the field radiated by the dipole layer (carrying an energy $U_c dz$). Therefore the field at z + dz has been rotated by an angle $d\varphi$ which is a fraction of φ equal to half of the energy radiated by the dipole layer to the total electromagnetic energy. This is written $d\varphi = \varphi (U_c dz)/2[(c/n)U_{em}\tau]$. The Verdet constant follows:

$$\rho = \frac{d\varphi}{Hdz} = \frac{g\beta}{\hbar} \frac{n}{c} \frac{U}{U_{\text{em}}} = \gamma A \tau^2$$
(4)

and is identical to the expression derived previously in the narrow-line case. Inspection of (3) and (4) shows clearly that large velocity reduction and large Faraday rotation are both related to large energy storage $U_C/U_{\rm em}$ in the resonant centers.

Giant and inverse Faraday effects are only two of many manifestations of this energy storage. Another is anomalous reflection at the boundaries of the resonant medium. The effect is purely electromagnetic in nature and directly related to the large reduction in group velocity. It is caused by an anomalous component of the magnetic field vector, which lies in the direction of the electric vector, and therefore does not contribute to energy flow, and whose envelope is the time derivative of the pulse envelope. All these effects provide additional techniques for the measurement of transparency. The giant Faraday effect may be the most attractive since large rotations can be achieved while applying only moderate magnetic fields.⁸ Measurement of the Verdet constant gives information about the spectral distribution function and the pulse length which is different from that obtained from a measurement of the pulse delay.

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- ²S. L. McCall and E. L. Hahn, Phys. Rev. Letters <u>18</u>, 908 (1967).

³C. K. N. Patel and R. E. Slusher, Phys. Rev. Letters <u>19</u>, 1019 (1967).

⁴The present discussion is limited to a resonant transition which can be treated as the superposition of two noninteracting two-level systems. If both σ_{-} and σ_{+} transitions to different upper-state Zeeman sublevels can be made from the same ground-state sublevel, single centers are excited to a superposition of states by a linearly polarized pulse. The lack of factorization of the resulting density matrix is not a trivial complication.

⁵P. S. Pershan, J. P. van der Ziel, and L. D. Malmstrom, Phys. Rev. <u>143</u>, 574 (1966).

⁶The success of our derivation is related to the fact that, in view of the slowly varying envelope approximation, the ratio of electric to magnetic field components in the 2π pulse is essentially the same as for the fields in the naked host.

⁷R. P. Feyman, F. L. Vernon, Jr., and R. W. Hellwarth, J. Appl. Phys. <u>28</u>, 49 (1957).

⁸In ruby, for the ⁴A₂ $(M_S = \pm \frac{1}{2}) \leftrightarrow {}^{2}E(\overline{E}) (M_{S'} = \pm \frac{1}{2})$ transition which was used in the first transparency experiment,² the constant $\gamma A = 2.5 \times 10^{17}$ rad/Oe cm sec², assuming 0.05% Cr³⁺ concentration. Assuming an inhomogeneous linewidth of the order of 0.1 cm⁻¹ we find $\rho = 5 \times 10^{-3}$ rad/Oe cm, a value so large that the effect could be very conveniently observed.