"Negative" Tensor Susceptibility in Media Exhibiting Population Inversion

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This paper describes the results of a theoretical and experimental investigation of the tensor dielectric susceptibility of a plasma in which some of the participating atoms exhibit population inversion. The measurement of the real part of the tensor dielectric susceptibility in the vicinity of an optical resonance has previously been very difficult due to the large absorption near resonance; however, by examining a resonance for which the levels exhibit population inversion we have obtained a direct measure of the real part of the susceptibility by observing the "negative" Faraday rotation, i.e., "negative" circular birefringence associated with the resonance. We also observed for levels with inverted populations the complement of the inverse Zeeman effect, that is, we observed a Zeeman splitting of the gain curve in the amplifying medium. The high-gain $5d[\frac{3}{2}]_1{}^0-6p[\frac{3}{2}]_1$ transition of atomic xenon at $2.026\,\mu$ in a helium-xenon mixture was used for these measurements. The observed results were consistent with the theory.

INTRODUCTION

We have observed large tensor dielectric susceptibility effects in the vicinity of an optical resonance between levels exhibiting population inversion. We have measured "negative" inverse Zeeman effect[1] and "negative" circular birefringence or Faraday rotation in a gaseous discharge medium as a function of applied magnetic field. In the case of uninverted populations, the real part of the tensor susceptibility is difficult to observe in the vicinity of resonance because of the strong absorption in the medium.[1,2] Even with media showing maser action (inverted populations), the negative absorption coefficients reported hitherto in gases have been too small (typically 0.10–0.15 per m) for practical investigation. However, with the discovery of large optical gain in a helium-xenon mixture[3] at $2.026\,\mu$ corresponding to the $5d[\frac{3}{2}]_1{}^0-6p[\frac{3}{2}]_1$ transition (Racah notation) of atomic xenon, a number of new experiments connected with investigation of various aspects of "negative" tensor susceptibility are now possible.

There have been studies of the effects of the scalar negative dispersion term in media not exhibiting inversion,[4,5] as early as 1932. The work presented clearly demonstrates the feasibility of extending this early work to include an examination in high resolution of both the real and the imaginary parts of the complex tensor dielectric susceptibility very close to and directly at resonance in media exhibiting a wide range of population differences including a high degree of inversion. This general approach is obviously applicable to solids and liquids as well as gases. It is also interesting to note that the perturbation of the dielectric susceptibility produced, for example, by Zeeman splitting, takes place very rapidly. This capacity for rapid perturbation coupled together with the capacity for monitoring the phase of the transmitted light at resonance suggests an excellent opportunity for an investigation of the time dependence of the optical dispersion process.[6] In addition, these techniques could be used to study the effects of a paramagnetic susceptibility[7,8] induced for example by selective optical pumping. The described effects can be used to produce rapid variations in the phase, frequency, amplitude and/or polarization of an intense beam of coherent light. This capacity should have general value in physical experiments as well as for modulation purposes.

THEORY

A simple theory which qualitatively predicts the observed results may be obtained from the complex scalar refractive index given by Bitter.[9] The real and imaginary parts of the index are, respectively, assuming only one isotopic species,

$$n_r \approx 1-\sum_j (\alpha_j\lambda/\pi^{1/2})[F(\omega_j)-\pi^{1/2}b\omega_j \exp(-\omega_j^2)],$$

$$n_i \approx (\alpha_j\lambda/2)\{\exp(-\omega_j^2)-(2b/\pi^{1/2})[1-2\omega_j F(\omega_j)]\}.$$

Assuming that all electronic transitions, denoted above by j, except one can be neglected, and denoting the magnetic sublevels by m, the gain coefficients for right and left circularly polarized light propagating along the field are

$$\alpha_\pm \approx \sum_m \alpha_{m,m\pm1}\{\exp(-\omega_{m,m\pm1}{}^2) - (2b/\pi^{1/2})[1-2\omega_{m,m\pm1}F(\omega_{m,m\pm1})]\}.$$

The corresponding refractive indices are

$$n_\pm \approx 1-\sum_m \frac{\alpha_{m,m\pm1}\lambda}{\pi^{1/2}} \times[F(\omega_{m,m\pm1})-\pi^{1/2}b\omega_{m,m\pm1}\exp(-\omega_{m,m\pm1}{}^2)].$$

[1] F. A. Jenkins and H. E. White, *Fundamentals of Optics* (McGraw-Hill Book Company, Inc., New York, 1957), 3rd ed.
[2] R. Fork, PhD thesis, Massachusetts Institute of Technology, 1962 (unpublished).
[3] C. K. N. Patel, W. L. Faust, and R. A. McFarlane (to be published).
[4] R. Ladenburg, Rev. Mod. Phys. **5**, 243 (1933).
[5] G. Breit, Rev. Mod. Phys. 4, 504 (1932); **5**, 91 (1933).
[6] O. Y. Savchenko, Opt. Spectr. (U.S.S.R.) **11**, 118 (1961).
[7] N. Bloembergen, P. S. Pershan, and L. R. Wilcox, Phys. Rev. **120**, 2014 (1960).
[8] H. G. Dehmelt, Phys. Rev. **105**, 1924 (1957).
[9] F. Bitter, Massachusetts Institute of Technology Research Laboratory of Electronics Tech. Rept. 292, 1955 (unpublished).

The various symbols are defined as follows:

$$\omega_{m,m'}\equiv[2(\ln 2)^{1/2}/\Delta\nu_D]\times[\nu-\nu_0+(H\mu_B/h)(mg-m'g')]\equiv\omega+\Delta\omega_{mm'},$$

$$\alpha_{m,m'}\equiv\beta_{m,m'}(\ln 2/\pi)^{1/2}(2\pi e^2 f/\Delta\nu_D m_0 c)(N_{m}'-N_m),$$

$$\sum_{m,m'}\beta_{m,m'}=1,$$

$$F(\omega)=\exp(-\omega^2)\int_0^\omega \exp(y^2)dy.$$

The constant b is given by $b=(\ln 2)^{1/2}\Delta\nu_N/\Delta\nu_D$. Here $\Delta\nu_N$ denotes the natural linewidth, $\Delta\nu_D$ denotes the Doppler linewidth, m_0 the electronic mass, f the oscillator strength, μ_B the Bohr magneton, H the magnetic field, and m' and m the magnetic substates of the upper and lower electronic states, respectively. The constant $\beta_{m,m'}$ represents the relative strength of the transition $m\rightarrow m'$ and N_m denotes the number of atoms per cc in magnetic sublevel m. It is noted that as $N_{m'}$ is made larger than N_m, a change of sign is introduced which, as well as changing the absorption into a gain, alters the sense of the birefringence and dichroism of the medium.

If the magnetic sublevels are unequally populated, the medium possesses a paramagnetic susceptibility and the dielectric susceptibility can be perturbed by both the Zeeman splitting, and by altering the level populations, for example, with a microwave field oscillating at the level splitting frequency. In gases the magnetic sublevels can be regarded as equally populated unless some special technique such as optical pumping is used to alter the Boltzmann distribution. In the work described the perturbation, therefore, arises almost entirely from Zeeman splitting.

Neglecting terms of the order of the ratio of the natural linewidth to the Doppler linewidth, the Faraday rotation per unit length can be written as

$$\chi=\frac{(n_+-n_-)}{2\lambda}=\tfrac{1}{2}\sum_m -[\alpha_{m,m+1}F(\omega_{m,m+1})-\alpha_{m,m-1}F(\omega_{m,m-1})].$$

A qualitative idea of the effects to be expected can be obtained by noting that the function $F(\omega)$ closely approximates the derivative of the Gaussian absorption curve.[9] For small fields the Faraday rotation is approximately $(\partial F/\partial\omega)\Delta\omega_{mm'}$ or the second derivative of a Gaussian times the Zeeman splitting and hence shows a maximum for incident light directly on resonance.

For $\nu\approx\nu_0$, χ is proportional to $F(\Delta\omega_{mm'})$, hence for incident light close to resonance the Faraday rotation plotted versus magnetic field closely resembles a dispersion curve. The order of magnitude of the rotation in xenon can be calculated from the g values $g=1.022$,[10] $g'\cong 1.0$,[11] the Doppler width $\Delta\nu_D\approx 200$ mc and the angular momenta of the two electronic states which both have $J=1$. For small fields, i.e., $\Delta\omega_{mm'}<0.2$, and assuming a gain of 4.5 dB per m as observed in reference 3, the calculated Faraday rotation is ≈ 0.39 deg/G m.

EXPERIMENT

The experimental arrangement consisted of a xenon oscillator followed by a xenon amplifier inside a solenoid and an analyzer placed between the amplifier exit and a detector. The oscillator served as a tunable source of nearly monochromaric linearly polarized light in the vicinity of the resonance to be investigated. Single frequency operation was insured by placing the oscillator mirrors sufficiently close so that only one longitudinal mode oscillated,[12] and the output was tuned over the Doppler linewidth by means of a magnetostrictive positioning device. The oscillator frequency was adjusted so as to coincide with the line center.

The expected variation in the observed intensity is obtained by considering a horizontally polarized beam of light passing through an amplifying medium of length l with gain α_0 per unit length and then through a polarizer oriented at θ with respect to the horizontal. The transmitted intensity is $I=I_0e^{\alpha_0 l}\cos^2(\theta+\chi)$, where χ is the Faraday rotation produced by the dispersing medium in the amplifier.

Two effects were observed, one arising from the variation in gain, the other from the Faraday rotation produced by the magnetic field. The gain coefficient exhibits symmetry about zero field, whereas the Faraday rotation exhibits an asymmetric dispersion curve shape. The two effects are easily separated by comparing ef-

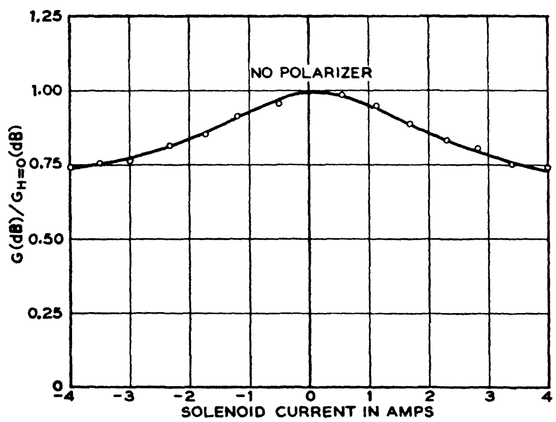

FIG. 1. G(dB)/$G_{H=0}$(dB) plotted as a function of solenoid current ($H=80$ G/A) in absence of polarizer.

[10] C. E. Moore, *Atomic Energy Levels*, National Bureau of Standards Circular No. 407 (U. S. Government Printing Office, Washington, D. C., 1958).

[11] Calculated.

[12] H. Kogelnik and C. K. N. Patel, Proc. I.R.E. (to be published).

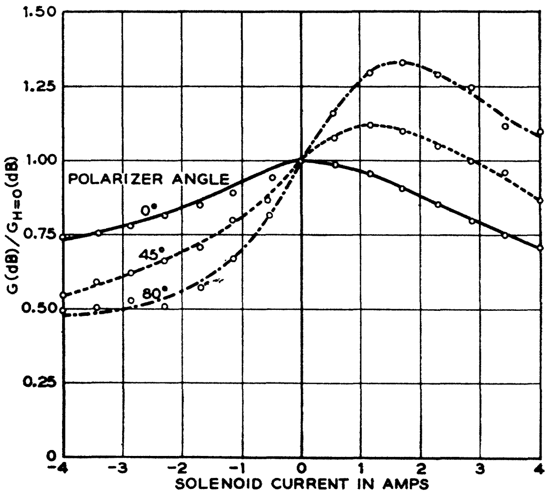

FIG. 2. G(dB)/$G_{H=0}$(dB) plotted as a function of solenoid current ($H=80$ G/A) for polarizer angles $\theta=0°$, 45°, and 80°.

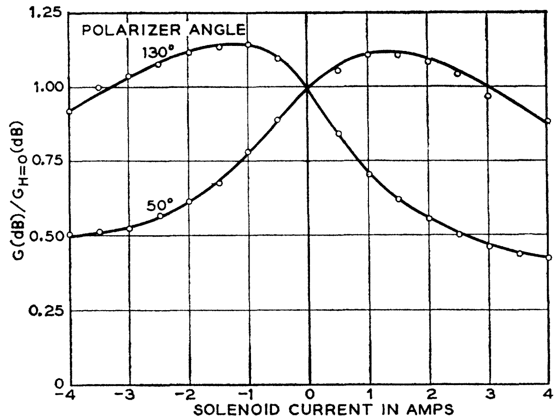

FIG. 3. G(dB)/$G_{H=0}$(dB) plotted as a function of solenoid current ($H=80$ G/A) for polarizer angles of $\theta=50°$ and 130°.

fective gain curves with the polarizer absent and with the polarizer present. Figure 1 shows curves taken in the absence of the analyzer. The symmetric dependence of the gain on magnetic field arising from the "negative" inverse Zeeman effect is apparent. In Fig. 2 the effective gain curve is plotted versus field for the case where an analyzer has been inserted with its axis at various angles to the polarization vector of the beam leaving the oscillator. Here the Faraday rotation and the analyzer cause a change in the net or effective gain of the combined amplifier-analyzer system clearly introducing an asymmetry. In Fig. 3 curves are given for the polarizer at 50° and 130° where the curves should be mirror images of each other. The slight lack of symmetry probably arises from an imperfect polarizer and changes in the discharge conditions.

We also observed a change in the sense of Faraday rotation on changing the sign of the term $(N_m-N_{m'})$, that is, on changing the gain into an absorption or vice versa. The relative populations were altered by varying the rf power coupled into the discharge. A polarizer was also inserted within the optical maser cavity and 100% amplitude modulation of the output was observed for an axial magnetic field oscillating at 1 kc/sec. This may be explained as the result of a time-dependent variation in the net gain produced primarily by Faraday rotation in the laser medium.

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