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# Polarization Rotation Induced by Resonant Two-Photon Dispersion

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A polarization-rotation effect is demonstrated which utilizes the dispersion associated with 3*S*-5*S* two-photon transitions in sodium vapor. A linearly polarized beam at $\nu_1$ is rotated by a circularly polarized beam at $\nu_2$ when $\nu_1+\nu_2$ is near the two-photon transition frequency. When combined with a polarizer a rapid optical shutter is obtained.

Recently there has been considerable interest in two-photon processes in atomic and molecular vapors.[1] Until now, however, most experiments have dealt only with the absorptive process, i.e., the imaginary part of the two-photon susceptibility. The dispersive part of the two-photon susceptibility can also produce strong effects and many new and important phenomena based on two-photon dispersion can be expected. We report here on one such phenomenon. We have made measurements of the polarization rotation which is produced by the dispersion associated with an *S*-to-*S* two-photon transition in an atomic vapor. This rotation effect is unique to the two-photon process and, unlike other effects of two-photon dispersion such as self-defocusing,[2] it has no single-photon analog.

A rotation of the polarization of a linearly polarized dye-laser beam of wavelength $\lambda_1$ (signal laser) is produced with a circularly polarized, control-laser beam of wavelength $\lambda_2$. The wavelengths are chosen such that the sum frequency of the two lasers is near but not equal to the 3*S* → 5*S* two-photon transition in atomic sodium. The magnitude and resonant behavior of the effect is found to be in good agreement with theory. When combined with a polarizer, a fast, optically controlled shutter or modulator is obtained.

Many phenomena which utilize the dispersion associated with single-photon transitions in atomic or molecular vapors have been previously studied. Transient and cw self-focusing effects have been investigated by Grischkowsky[3] and by Bjorkholm and Ashkin.[4] Gibbs, Churchill, and Salamo[5] have reported Faraday-rotation angles in excess of 180° which were produced in sodium vapor in a magnetic field of 1 kG, and resonant birefringence due to optically induced level shifts has been reported by Bonch-Bruevich, Kostin, and Khodovoi.[6] In addition the large dispersion available with near-resonant atomic and molecular transitions has recently been used to produce pulse compression and the conversion of cw light into pulses by Grischkowsky,[7] by Loy,[8] and by Bjorkholm, Turner, and Pearson.[9]

A major limitation of phenomena arising from single-photon dispersion is the requirement of a near-resonant atomic or molecular transition. This restriction is considerably relaxed when one uses two-photon transitions. In a two-photon transition we can picture the control-laser beam at frequency $\nu_2$ as inducing the atom to exhibit an absorption resonance at $\Omega-\nu_2$, where $\Omega$ is the two-photon transition frequency. From the Kramers-Kronig relationships one finds a dispersion associated with this induced absorption resonance. The resonance frequency of this induced absorption can be adjusted by adjusting the control-laser frequency, $\nu_2$, and hence the need for a chance coincidence with atomic transition frequencies is removed.

The polarization-rotation effect reported here is a result of the selection rules for two-photon absorption. In particular, the selection rules[10] for *S*-to-*S* transitions in atomic vapors are such that if circularly polarized photons are used, the two absorbed photons must have opposite senses of circular polarization. Therefore, if the control-laser beam at $\nu_2$ is circularly polarized, a two-photon, $S$-to-$S$ transition will affect only one of the two circularly polarized components of the linearly polarized signal beam at $\nu_1$. By correctly adjusting $\nu_2$ such that $\Omega - \nu_2$ is nearly equal to $\nu_1$, the two-photon dispersion can produce a considerable phase shift for the one circularly polarized component of $\nu_1$ while avoiding significant absorption of either beam. The result is a simple rotation of the direction of polarization of the beam at $\nu_1$. The rotation is similar to that obtained in the Faraday effect; however, a magnetic field is not required.

The rotation angle in radians is given by

$$\Phi = \frac{16\pi^3 lN}{h^3c^2}\left|\sum_n \mu_{gn}\,\mu_{ne}\left(\frac{1}{\nu_1-\nu_n}+\frac{1}{\nu_2-\nu_n}\right)\right|^2 \times \frac{\nu_1}{\Omega-(\nu_1+\nu_2)}I,$$

where $I$ is the intensity of the control laser, $l$ is the length of the vapor cell, $N$ is the vapor density, $\mu_{gn}$ and $\mu_{ne}$ are dipole matrix elements for circularly polarized light, and the sum is taken over all intermediate states with energies $E_n = h\nu_n$. In deriving this expression it is assumed that the atomic and laser linewidths are small compared to $|\nu_1-\nu_n|$, $|\nu_2-\nu_n|$, and $|\Omega-(\nu_1+\nu_2)|$. Under these assumptions the speed of this rotation phenomenon is limited only by the temporal properties of the control-beam pulse.

In Fig. 1 we show a schematic diagram of the experimental setup. The control and signal beams were generated by two dye lasers which were simultaneously pumped with a single $N_2$ laser. The bandwidth of each laser was approximately 0.2 Å. After passing through appropriate polarizers the two beams were combined with a beam splitter and passed collinearly through approximately 5 cm of sodium vapor at a density of $3.2\times10^{14}/\text{cm}^3$ (~ 0.01 Torr). The vapor was contained with 7 Torr of argon buffer gas. The control beam was focused to about a 1-mm-diam spot, while the signal beam was focused to approximately 0.2 mm so that there would be a fairly uniform rotation of the signal-beam polarization. At the location of the sodium vapor cell, the total peak powers of the control beam and signal beam were $1.5\times10^4$ W (peak power density $1.5\times10^6$ W/cm$^2$) and 40 W (peak power density $5.5\times10^4$ W/cm$^2$), respectively. After passing through the vapor, the signal beam was isolated with a filter and detected with a fast photodiode and a Tektronix 7904 oscilloscope. The combined response time was less than 1 nsec. The signal polarization was analyzed with a Glan polarizer. The control-laser pulse had a duration (full width at half-maximum) of 6.5 nsec while the signal-laser pulse width was 4.5 nsec. The optical paths from these lasers to the vapor cell were arranged such that the signal-laser pulse arrived within the control-laser pulse. All measurements of rotation angle and transmission were made at the peak of the control-laser pulse.

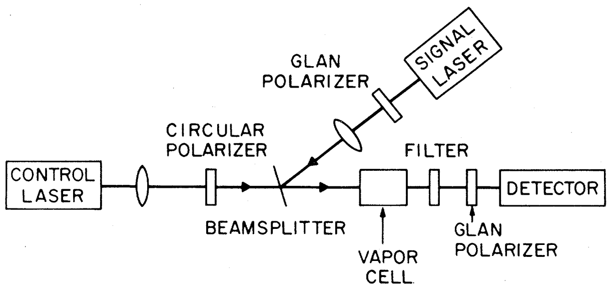

FIG. 1. Experimental setup.

Because of the limited amount of control-laser power available in our experiment, large rotation angles (angles ~ 90°) could only be obtained by adjusting the wavelengths of the two lasers for (1) strong resonant enhancement[11] of the two-photon transition by tuning the signal beam close to the $3S_{1/2}$-$3P_{3/2}$ or $3S_{1/2}$-$3P_{1/2}$ intermediate-state resonance and (2) strong dispersion by tuning the control laser such that the sum frequency was close to the $3S_{1/2}$-$5S_{1/2}$ two-photon resonance. In principle, sufficient control-laser power could produce large rotation angles for arbitrary mistuning from these resonances. Self-focusing and self-defocusing effects due to both the single-photon[3] and the two-photon[2] transitions were clearly evident for tuning close to either the single- or the two-photon transition. These effects were a power-handling limitation in our experiment.

The dependence of rotation angle on control-beam intensity is shown in Fig. 2. The signal was tuned at 5891 Å and the control laser at 6159 Å, i.e., 1 Å from both the $3P_{3/2}$ intermediate-state resonance and the $3S \rightarrow 5S$ two-photon resonance. The angle is seen to be linearly proportional to the control-laser intensity with a slope of $3.1\times10^{-5}$ deg/(W/cm$^2$). In view of the uncertainties involved in the measurements, this value is in good agreement with a calculated $1.6\times10^{-5}$ deg/(W/cm$^2$). The effects of self-focusing or self-defocusing have not been taken into ac-

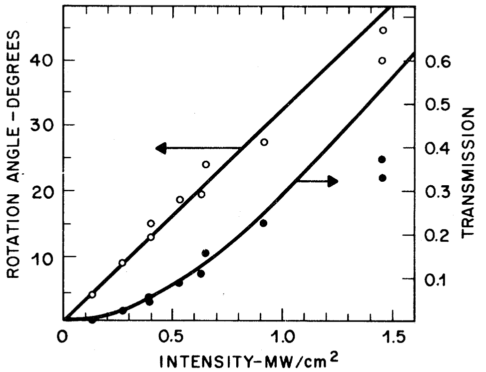

FIG. 2. Intensity dependence of rotation angle and transmission through crossed polarizer. Control and signal dye lasers tuned such that the mistuning from both the single-photon intermediate-state resonance and the two-photon resonance is 1 Å.

count in our calculation.

It is interesting to compare the rotation angle or equivalently the phase retardation which is measured here with that obtained with the electronic optical Kerr effect in glass. The phase retardation of $3.1\times10^{-5}$ deg/(W/cm$^2$) for a path length of 5 cm corresponds to an optical Kerr coefficient, $n_{2B}$, of $2.3\times10^{-10}$ esu. This value can be compared to that in glass of $2\times10^{-13}$ esu.[12] The large increase in the bulk nonlinear coefficient over that of glass is achieved in spite of a vapor density 7 to 8 orders of magnitude less than for solids and is a consequence of the resonant enhancement of the two-photon dispersion.

Also shown in Fig. 2 is the transmission through the Glan polarizer whose direction was crossed to that of the original signal polarization. The line drawn through the data points is $\sin^2\Phi$. Again there is good agreement between experiment and theory. The deviation is probably due to the finite bandwidth of the lasers resulting in different rotation angles for different spectral components, nonuniformity in the intensity profile of the control-laser beam, and the effects of self-focusing. The maximum transmission shown in Fig. 2 is about 40%; however, transmission up to 70% has been observed with this "shutter" in the open condition and less than 0.1% in the "closed" position. The latter was limited only by the quality of the polarizers. Because of the longer duration of the control-laser pulse relative to the signal-laser pulse, the rotation experienced by the signal pulse was nearly constant in time, and the signal which passed through the crossed polarizer was only slightly distorted. Its width showed a slight decrease from 4.5 to 4.0 nsec.

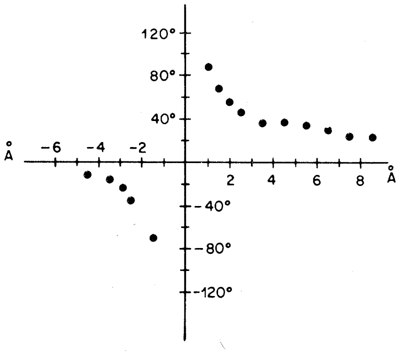

FIG. 3. Dependence of rotation angle on control-laser wavelength, in terms of mistuning from exact two-photon resonance. Signal laser tuned at 5890.6 Å and control-laser intensity is about $1.3\times10^6$ W/cm$^2$.

In Fig. 3 the effect of tuning the control-laser wavelength is shown. The signal laser is tuned to 5890.55 Å. The resonant character of the observed dispersion is clearly evident and in good qualitative agreement with theory; however, the data show a reproducible asymmetry about the wavelength of exact two-photon resonance. The rotation angles on the short-wavelength side of resonance are smaller than those obtained for an equal mistuning on the long-wavelength side. This asymmetry probably results from self-focusing effects. Both the two-photon and the single-photon transitions lead to self-defocusing for short wavelengths while on the long-wavelength side the two-photon contribution and single-photon contribution tend to cancel. More detailed experiments investigating these effects are in progress.

In the region near exact two-photon resonance the absorption of one circularly polarized component of the linearly polarized signal beam is so strong that the output is nearly 100% circularly polarized. The transmission through the polarizer is therefore primarily a result of this remaining component, and we observed no preferential direction of linear polarization.

The polarization-rotation effect described here is one of a new class of phenomena which utilize two-photon dispersion. It is an electronic effect whose speed is limited only by the temporal properties of the control-laser pulse, and it is produced in gas-phase media. Gases possess the advantageous properties of tolerance to high optical power densities, of transparency over wide ranges of the electromagnetic spectrum, and of ease of scaling to arbitrary physical dimensions.

By choosing the atomic system and the frequency of the control laser appropriately, it should be possible to control beams from the far-infrared to the vacuum ultraviolet. The rotation effect also provides a sensitive technique for measuring two-photon cross sections. Extensions of this work to other schemes which use the two-photon dispersion to provide pulse compression and reshaping are presently under investigation.

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# Magnetic Braiding Due to Weak Asymmetry*

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Magnetic surfaces for a plasma with a helical current perturbation $\sim \epsilon^2$ are destroyed by toroidal effects or by a second current perturbation, of incommensurate helicity, and the behavior of magnetic field lines becomes stochastic in layers of relative width $\epsilon^{-l} \times \exp(-\pi/2\epsilon)$, where $l \simeq 2|m_1/m| + 1$ with $m$ and $m_1$ the azimuthal mode numbers of the original helical field and of the perturbation.

This work considers how the magnetic surfaces for a tokamak discharge are affected by helical perturbations of the plasma current. Such current perturbations are known to be associated with resistive modes,[1,2] and substantial experimental evidence has been offered[3] for the occurrence of magnetic islands[4] associated with nonlinear tearing instabilities.[5,6] In this paper we show that the existence of two such modes with different helicity *or* the effect of toroidal geometry on a single such mode leads to the destruction of magnetic surfaces. The resultant stochastic wandering or "braiding" of the magnetic lines can produce collisionless radial heat transport, enhanced current penetration, and radial particle transport, and may change the inductance for toroidal plasma current flow so that sudden onset of braiding would produce negative or positive spikes in the loop-voltage signal.[7]

The mechanism of magnetic-surface destruction[8] was first investigated in two classic papers on magnetic irregularities,[9,10] where it was demonstrated that a spectrum of overlapping resonances produces stochastic wandering of the magnetic field lines. In this work we start with a field of helical symmetry and exact magnetic surfaces which exhibits a single set of primary islands, i.e., a single resonance at some $r = r_0$ between the helical variation and the rotational transform $\iota(r)$. Weak asymmetry is introduced via a first-order magnetic perturbation of different helicity which might be due to toroidal effects or to the presence of a second magnetic resonance at $r = r_1$. The incommensurate perturbation is found to produce little *secondary* islands which appear wherever the Fourier components of the perturbation resonate with the *local* transform, $\omega(k)$, Eq. (4), within the *primary* islands.