PHYSICAL REVIEW A **VOLUME 40, NUMBER 6** SEPTEMBER 15, 1989

Energy transfer between laser beams propagating through an atomic vapor

Mark T. Gruneisen,* Kenneth R. MacDonald,[†] Alexander L. Gaeta, and Robert W. Boyd Institute of Optics, University of Rochester, Rochester, New York 14627

Donald J. Harter

Allied Signal, Inc., Columbia Road and Park Avenue, Morristown, New Jersey 07960 (Received 17 April 1989)

We show that it is possible to transfer energy efficiently between two laser beams of comparable intensity by means of the near-resonant nonlinear response of an atomic vapor. The coupling is due to stimulated Rayleigh scattering and occurs when the frequency difference between the two beams is approximately the inverse of the lifetime of the excited atomic state. We have measured the coupling between two beams from an Alexandrite laser tuned near the $4^{2}S_{1/2} \rightarrow 4^{2}P_{3/2}$ potassium transition and found that one of the transmitted beams can contain up to 85% of the total incident energy.

Laser beams of extremely high intensity can be obtained by coherently combining the output beams of a large number of independent lasers. In this Rapid Communication, we examine a new mechanism for laser beam combining based on the nonlinear optical response of a resonantly excited atomic vapor. Atomic vapors are particularly appealing for this purpose because of their highly nonlinear response. Unlike most previous studies of the resonant interaction of laser radiation with an atomic system, we consider the case in which the atom interacts with two laser fields, each of which is sufficiently intense to saturate the atomic transition.

The nonlinear response of a strongly driven atomic system has attracted great interest. Mollow' has shown that the absorption spectrum experienced by a *weak* probe wave in the presence of an intense, near-resonant pump wave consists of three features, two of which can lead to amplification of the probe wave. One of these gain features occurs when the probe frequency is within $f=1/T_1$ of the pump laser frequency (where T_1 is the atomic excited-state lifetime) and is a form of stimulated Rayleigh scattering. The other gain feature occurs when the frequency of the probe wave is detuned from that of the pump wave by the generalized Rabi frequency associated with the atomic response and occurs due to stimulated three-photon scattering.^{2,3} The existence of these two gain features was verified by means of an experiment performed using a sodium atomic beam.⁴ We have recently performed a similar experiment using a sodium atomic vapor⁵ where much larger atomic number densities, and hence a stronger nonlinear coupling, could be achieved. With proper choice of pump laser intensity and detuning from the Doppler broadened atomic resonance, we were able to achieve a 38-fold increase in the intensity of the probe wave in a 7-mm interaction length by means of the feature near the Rabi sideband.

In this Rapid Communication, we consider the coupling between two laser beams for the case in which both waves are strong and of comparable intensity. In particular, we are interested in determining the conditions under which efficient transfer of energy between the two beams is possible. We have solved the Maxwell-Bloch equations describing the propagation of two intense laser fields through a vapor of two-level atoms. The details of this calculation will be given elsewhere.⁶ We find that the intensities of the two waves vary spatially according to

$$
\frac{dI_j}{dz} = -\alpha_j (I_1, I_2) I_j , \qquad (1)
$$

where I_j is the intensity of the jth wave and α_j is an effective, intensity-dependent absorption coefficient given by

$$
\alpha_j(I_1, I_2) = -N\sigma_0\Lambda_j w_0\{1 - I_{3-j}[Re(L_1F)]\},
$$

$$
\times (-1)^{j+1}\Delta_j T_2[M(L_1F)]\},
$$

(2)

where N is the number density, σ_0 is the weak-field linecenter absorption cross section, $\Lambda_j = [1 + (\Delta_j T_2)^2]^{-1}$ is a Lorentzian detuning factor, w_0 is the spatially averaged, steady-state population inversion in the presence of the two waves, $\Delta_i = \omega_i - \omega_0$ is the detuning of the *j*th beam from the atomic resonance frequency ω_0 , and F is a continued fraction given by

$$
F = \frac{1}{1 + \Phi_1 - \frac{(L_2)^2 I_1 I_2}{1 + \Phi_2 - \frac{(L_3)^2 I_1 I_2}{1 + \Phi_3 - \cdots}}},
$$
(3)

where $\Phi_n = M_{1n}I_1 + M_{2n}I_2 - in\delta T_1$ and $\delta = \omega_2 - \omega_1$. The. terms L_n and M_{in} are given by

$$
L_n = \frac{1}{2I_s} \{ [1 - i(\Delta_1 + n\delta)T_2]^{-1} + [1 + i(\Delta_2 - n\delta)T_2]^{-1} \}
$$
\n(4a)

and

$$
M_{jn} = \frac{1}{2I_s} \{ [1 - i(\Delta_j + n\delta)T_2]^{-1} + [1 + i(\Delta_j - n\delta)T_2]^{-1} \},
$$
\n(4b)

where $I_s = c\hbar^2/8\pi |\mu|^2 T_1 T_2$ is the line-center saturation intensity and μ is the atomic dipole matrix element.

Several previous workers have shown that the solution of the Bloch equations in the presence of two strong fields can be represented in terms of a continued fraction solu- tion ; however, we are unaware of any previous workers who have considered the propagation of two intense interacting fields through an atomic vapor.

Figure ¹ shows how the predicted spectrum of the effective absorption coefficient a_2 experienced by the wave at frequency ω_2 is modified as its intensity I_2 is increased. For convenience we refer to this wave as the probe wave and to the wave at frequency ω_1 as the pump wave, even though the interaction is symmetric between the two waves. In all three plots, we consider the case of a radiatively broadened medium with $T_2/T_1 = 2$, a pump wave detuning from resonance of $\Delta_1 T_1 = -25$, and a pump wave intensity of 5×10^3 in units of the saturation intensity. In Fig. 1(a) we consider the case of a weak probe wave, in which case our predictions reduce to those of the theory of Mollow.¹ The left-most feature occurs at the Rabi sideband of the pump frequency [i.e., at $\omega_2 - \omega_1$ α' where $\Omega' = (I_1/I_s T_1 T_2 + \Delta_1^2)^{1/2}$, and arises due to stimulated three-photon scattering. The central feature of the spectrum is due to stimulated Rayleigh scattering. The effects of these gain features are known to lead to Rabi sideband generation,^{3,8} conical emission,⁹ paramet Rabi sideband generation,^{3,8} conical emission,⁹ paramet·
ric instabilities,¹⁰ and parametric oscillation.¹¹ The right· most feature in this spectrum is the ac-Stark-shifted atomic resonance, and always leads to attenuation of the probe beam. In Figs. $1(b)$ and $1(c)$ we show how the predicted spectra are modified as the intensity of the probe beam is increased to 2% of the pump intensity in Fig. 1(b)

FIG. 1. Probe-wave absorption coefficient vs probe-pump detuning for several values of the probe-pump intensity ratio, for the case $I_1 = 5 \times 10^3 I_s$, $\Delta_1 T_1 = -25$, and $T_2/T_1 = 2$.

and to be equal to the pump intensity in Fig. 1(c). In each case, the magnitudes of the Rabi sidebands are seen to decrease as new features appear at subharmonics of the Rabi frequency. We also see that the Rayleigh feature, which is almost imperceptible in Fig. 1(a), becomes the dominant feature in Fig. 1(c).

In order to make predictions relevant to the case of an atomic vapor, we include the effects of Doppler broadening of the atomic resonance frequency by performing a numerical average of the absorption coefficients over a Maxwellian distribution of atomic velocities. In the experimentally realistic limit in which the Doppler linewidth is much greater than the homogeneous linewidth and in which the beams are nearly copropagating, we find that the fundamental and subharmonic Rabi resonances are largely washed out by the effects of atomic motion, but that the Rayleigh resonance remains and is relatively unaffected. The reason for this behavior is that the positions of the Rabi resonances depend upon the atomic velocity through the dependence of the generalized Rabi frequency on the detuning of the laser from resonance; conversely, the Rayleigh resonance is relatively unaffected by atomic motion because it is always centered on the pump laser frequency. Hence, we have used the Rayleigh resonance in our experimental studies of laser energy transfer.

We calculated the predicted transfer efficiency, defined as the fraction of the total input power that is contained in one of the output beams, by numerically integrating the set of Eq. (1). For sufficiently high input intensities and sufficiently long interaction path lengths, we find that the transfer efficiency can exceed 95% for the case of equal input intensities. The transfer efficiency is maximized by setting δ approximately equal to $1/T_1$ (that is, by utilizing the peak of the Rayleigh resonance) and setting Δ_1 equal to $\frac{1}{2}$ of the negative of the generalized Rabi frequency associated with one of the input waves.

We have investigated these predictions using the experimental setup shown in the inset to Fig. 2. The output of

FIG. 2. Energy-transfer efficiency, defined as the ratio of the output energy of the probe wave to the total input energy, as a function of the pump-wave detuning from resonance. The experimental setup is shown in the inset (a.o.m. is the acoustooptic modulator, K is the potassium vapor cell). The pump and probe waves have equal input energies of 20 μ J, the detuning of the probe wave from the pump wave is $\delta/2\pi = 3$ MHz, and the temperature of the body of the potassium cell is 220° C. The solid curve gives the best theoretical fit to the experimental data.

an Alexandrite laser operating in a single transverse and single longitudinal mode at a wavelength of 760 nm and producing up to 250 μ J of energy in a 500-ns pulse is passed through a variable attenuator and split into two beams by two acousto-optic modulators operating at slightly different frequencies. These beams are adjusted to have equal pulse energies and equal diameters of \sim 1 mm. These beams are allowed to intersect at an angle of 0.60° in a 7-mm-long potassium vapor cell containing no buffer gas and operating at a temperature of up to 250° C. The radiative lifetime of this transition is $T_1 = 25$ ns and the dipole dephasing time is calculated using the formu- $\ln^{12} T_2 = 1/(1/2T_1 + 2.07 \times 10^{-7})$. Some of our experimental results are shown in Fig. 2, where the energytransfer efficiency (that is, the ratio of the energy in one of the output beams to the total input energy) is plotted as a function of the detuning of the pump wave from the $4^{2}S_{1/2} \rightarrow 4^{2}P_{3/2}$ transition frequency. The frequency difference between the two waves $\delta/2\pi$ was held fixed at 3 MHz, which was the value that gave the best energy transfer. The input laser pulses each contained 20 μ J of energy. The solid curve shows the best theoretical fit to the data. In obtaining this curve, we take the intensity of each wave to be 33 kW/cm² corresponding to I_1/I_s $=I_2/I_s = 1 \times 10^7$. We also assume that the temperature of the atomic vapor is 175° C, which corresponds to the ratio T_2/T_1 being equal to 1.3, and the absorption path length

100-' $(%)$ 85%g 2 $\frac{1}{5}$ D. $\frac{D}{D}$ յե τ . $~\cdot$

 Ω

total input energy (μJ)

FIG. 3. Energy-transfer efficiency for the probe and pump waves as functions of the total input energy. The pump and probe waves have equal input energies, the probe-pump detuning is $\delta/2\pi = 3$ MHz, the detuning of the pump wave from resonance is $\Delta_1 = -3.8$ GHz, and the temperature of the body of the cell is 250'C. For a wide range of input energies, the transfer efficiency for the probe wave exceeds 80%. The solid curve gives the best theoretical fit to the experimental data.

and 3, it was necessary to assume that the laser intensity was 6 times higher than the measured value and that the vapor temperature was 45° C lower than that measured at a reference point on the outside of the potassium cell. We believe that these discrepancies may be due to our theoretical assumption that the beams were of monochromatic plane waves, due to optical pumping effects which could make the effective atomic number density lower than the actual density, due to self-defocusing effects, or due to temperature gradients within the vapor cell.

In conclusion, we have shown that efficient energy coupling $($ > 85%) between two laser beams can occur in an atomic vapor cell over a wide range of input intensities. This technique appears to be a promising one for combining beams of very high energy since the transfer efficiency is predicted to be nearly equal to unity for input intensities and interaction path lengths somewhat larger than those used in the present experiment.

This research was supported by the Office of Naval Research Contract No. N00014-86-K-0746 and the sponsors of the New York State Center for Advanced Optical Technology. One of us (M.T.G.) acknowledges support by the U.S. Army Office of University Research Initiative.

Present address: Air Force Weapons Laboratory/AROM, Kirtland Air Force Base, NM 87117.

We have also performed experiments aimed toward maximizing the energy-transfer efficiency. The maximum coupling was found to occur for a pump-probe detuning of $\delta/2\pi = 3$ MHz, a pump detuning of $\Delta_1 = -38$ GHz, and with the potassium cell held at a temperature of 250° C. The results of the study are shown in Fig. 3, where we plot the transfer efficiency for each wave as a function of the total input energy. At very low input energies, essentially no coupling between the beams takes place, and each beam exists with approximately half of the total incident energy. However, as the total input energy is increased to above 100 μ J, a significant amount of energy transfer occurs, leading to as much as 85% of the total energy being contained in one of the output beams. The solid line is a theoretical curve obtained by assuming that the beam diameters were 0.4 mm and that the temperature of the atomic vapor was $205\,^{\circ}$ C. In order to obtain good agreement between theory and experiment as shown in Figs. 2

- tPresent address: Rockwell Power Systems, 2021 Girard S.E., Albuquerque, NM 87106.
- ¹B. R. Mollow, Phys. Rev. A 5, 2217 (1972).

 $N\sigma_0 L$ being equal to 5.6 \times 10⁴.

- $2M$. Sargent III, Phys. Rep. 43, 223 (1978); S. E. Schwartz and T. Y. Tan, Appl. Phys. Lett. 10, 4 (1967); L. W. Hillman, R. W. Boyd, J. Krasinski, and C. R. Stroud, Jr., J. Opt. Soc. Am. B 1, 73 (1984).
- ³R. W. Boyd, M. G. Raymer, P. Narum, and D. J. Harter, Phys. Rev. A 24, 411 (1981).
- ⁴F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. 3\$, 1077 (1977).
- 5M. T. Gruneisen, K. R. MacDonald, and R. W. Boyd, J. Opt. Soc. Am. B 5, 123 (1988).
- M. T. Gruneisen, K. R. MacDonald, A. L. Gaeta, R. W. Boyd, and D. J. Harter (unpublished).
- ⁷R. Gush and H. P. Gush, Phys. Rev. A 6 , 129 (1972); N. Tsukada, J. Phys. Soc. Jpn. 46, 1280 (1979); A. M. Bonch-Bruevich, T. A. Vartanyan, and N. A. Chigir, Zh. Eksp. Teor. Fiz. 77, 1899 (1979) [Sov. Phys. JETP 50, 901 (1979)];G. I. Toptygina and E. E. Fradkin, Zh. Eksp. Teor. Fiz. 82, 429 (1982) [Sov. Phys. JETP 55, 246 (1982)];G. S. Agrawal and N. Nayak, J. Opt. Soc. Am. B 1, 164 (1984); L. W. Hillman, J. Krasinski, K. Koch, and C. R. Stroud, Jr., ibid. 2, 211 (1985).

pump

I 200

- ⁸D. J. Harter, P. Narum, M. G. Raymer, and R. W. Boyd, Phys. Rev. Lett. 46, 1192 (1981).
- 9D.J. Harter and R. W. Boyd, Opt. Lett. 7, 491 (1982).
- ¹⁰S. L. McCall, Phys. Rev. A 9, 1515 (1974); R. Bonifacio and L. A. Lugiato, Lett. Nuovo Cimento 21, 510 (1978); A. C. Tam, Phys. Rev. A 19, 1971 (1979); L. W. Hillman, R. W. Boyd, and C. R. Stroud, Jr., Opt. Lett. 7, 426 (1981); S. Hendow and M. Sargent III, Opt. Commun. 40, 385 (1982); I.
- Bar-Joseph and Y. Silberberg, Phys. Rev. A 36, 1731 (1987); G. Khitrova, J. F. Val1ey, and H. M. Gibbs, Phys. Rev. Lett. 60, 1126 (1988).
- ¹¹D. Grandclement, G. Grynberg, and M. Pinard, Phys. Rev. Lett. 59, 44 (1987).
- ¹²R. B. Miles and S. E. Harris, IEEE J. Quantum Electron. QE-9, 470 (1973).