Observation of electromagnetically induced phase matching

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We report the observation of electromagnetically induced phase matching in collisionally broadened Pb vapor. At a critical intensity at which the Rabi frequency of a dressing 1064-nm laser overcomes the Doppler broadening of the vapor, the generated four-frequency-mixing signal at 283 nm increases in a steplike manner by a factor of 59.

By applying a strong orthogonally polarized 1064-nm laser, Field *et al.*[1] observed a large increase in the transparency of collisionally broadened Pb vapor to an applied near-resonant 283-nm laser beam. This transparency results from a Fano-like interference in the absorption to the dressed states, which are created by a 1064-nm laser (Fig. 1, inset). It has been predicted[2] that this interference will be destructive in both the real and imaginary parts of the 283-nm linear susceptibility and will be constructive in the nonlinear susceptibility for the generation of this wavelength by four-frequency mixing.

In this Letter we show how the destructive interference in the real part of the 283-nm susceptibility may be used to electromagnetically phase match a four-frequency-mixing process. With weak intensities of the 1064-nm laser, the detuning of 283 nm from the Pb resonance transition is 6 cm^{-1} (Fig. 1), and, for the conditions of this experiment, the coherence length for its generation is 0.26 mm and the absorption depth is 50 mm. Since the coherence length is much smaller than the absorption depth, the effect of electromagnetically induced phase matching dominates over electromagnetically induced transparency in this experiment. In experiments in which both ω_c and ω_d (Fig. 1) are exactly or nearly on resonance, the effect of electromagnetically induced transparency will be dominant.

As the intensity of the 1064-nm laser is increased, the energies of the dressed atomic states increase and decrease, respectively, and cause the real part of the susceptibility to have a steep (positive) slope with a zero at the sum of bare state $|2\rangle$ and 1064 nm.[3,4] In the absence of inhomogeneous broadening and for zero linewidth of the $|1\rangle-|2\rangle$ transition, any finite intensity of the 1064-nm laser causes the four-frequency process to be exactly phase matched at a frequency equal to or extremely close to the zero of the real part of the linear susceptibility. When the $|1\rangle-|2\rangle$ transition linewidth is broadened, either homogeneously or inhomogeneously, there is a critical 1064-nm field strength at which this broadening is overcome. At this critical intensity the medium becomes electromagnetically phase matched, and one expects a steplike increase in the mixing efficiency of the four-frequency process.[2] If the 1064-nm laser were exactly on resonance, this critical intensity would occur when the Rabi frequency exceeds the $|1\rangle-|2\rangle$ transition linewidth. In this experiment, the 1064-nm laser is detuned from resonance by 6 cm^{-1}, and the critical intensity occurs when the square of the Rabi frequency is approximately equal to the product of the $|1\rangle-|2\rangle$ transition linewidth and the detuning.

We generate a 283-nm beam by the four-frequency-mixing process of Fig. 1 and observe the intensity at 283 nm as a function of 1064-nm laser intensity. At a critical intensity we observe a steplike increase in the generated signal at 283 nm, with an enhancement of 59. We note, though, as is discussed below, that this enhancement is far less than that which is predicted for an experiment with monochromatic lasers and a cell that is optically thick at 283 nm; for such an ideal experiment an enhancement of approximately 6200 is expected.

Before proceeding we review earlier research. Hakuta *et al.*[5] used atomic hydrogen and a dc field to demonstrate a resonantly enhanced nonlinear susceptibility, together with reduced loss at a generated wavelength of 121.6 nm. Tewari and Agarwal[6] suggested that dense media may be phase matched by applying a laser field that is not a part of the four-frequency-mixing chain and that therefore results in a destructive interference in both the linear and nonlinear susceptibilities but may still, in certain cases, produce comparable efficiency. Hahn *et al.*[7] used a destructive interference between autoionizing states in Zn to generate a 104.8-nm beam.

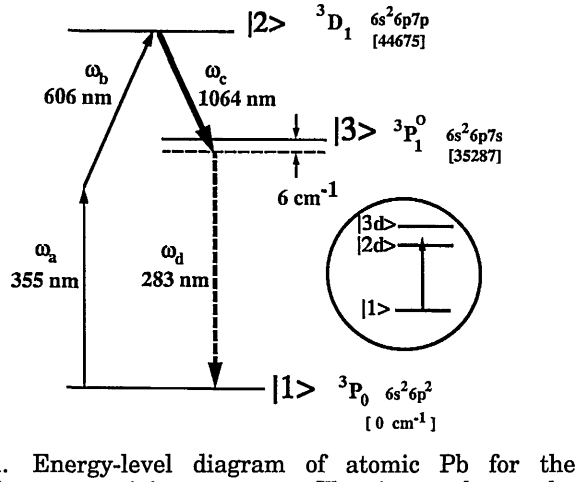

Fig. 1. Energy-level diagram of atomic Pb for the four-frequency-mixing process. The inset shows the dressed atomic states as seen by the generated 283-nm light.

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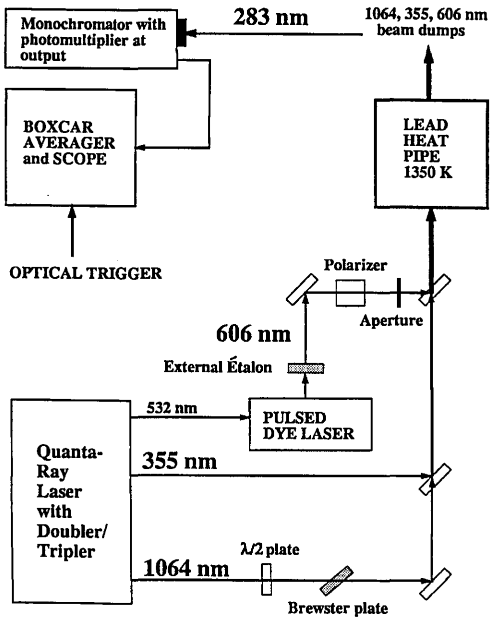

Fig. 2. Schematic of the experimental apparatus.

A schematic of our experimental setup is shown in Fig. 2. The 1064-nm Nd:YAG laser is injection seeded by a diode-pumped monolithic nonplanar ring oscillator (Lightwave Electronics Model S-100) and has a single-mode transform-limited FWHM pulse length of 10 ns. This laser is frequency tripled to provide 355-nm radiation and is also frequency doubled to provide the pumping wavelength for the tunable 606-nm radiation. The 606-nm radiation is obtained from a pulsed dye laser (Quanta-Ray Model PDL1) with an intracavity étalon and has a measured linewidth of 0.08 cm^{-1} and a pulse length of 4 ns. An external étalon reduces this linewidth to 0.03 cm^{-1}.

The Pb vapor cell is made of tantalum, is heated by a 9-cm-long molybdenum heater, and is run at a He buffer gas pressure of 3.5 Torr. As required by the selection rules for the Pb transitions, the applied 606-nm radiation is orthogonally polarized to the 1064- and 355-nm beams (both horizontally polarized), thereby generating vertically polarized 283-nm light. To ensure that the area (50 mm^2) of the 1064-nm beam is larger than that of the generated beam, the 606-nm laser is passed through a 5-mm^2 aperture. At the input of the Pb heat pipe, the per-pulse energies and power densities of the 1064-, 355-, and 606-nm lasers are 120 mJ, 325 μJ, and 0.25 μJ and 24 MW/cm^2, 130 kW/cm^2, and 1.25 kW/cm^2, respectively.

The generated 283-nm light, as measured before the monochromator, has a per-pulse energy of approximately 0.1 nJ. It is detected by a Hamamatsu R1463 photomultiplier, averaged over 30 samples with a boxcar integrator, and displayed on a digital multimeter. It is observed that the 283-nm signal disappears if the polarization of the 606-nm laser is changed from vertical to horizontal and is greatly reduced if the Quanta-Ray seeder is disabled.

To demonstrate electromagnetically induced phase matching we observe the detected power at 283 nm as a function of the 1064-nm power density. The 606- and 355-nm power densities are held fixed at 1.25 and 130 kW/cm^2, respectively. Figure 3 shows the ratio of the detected power density at 283 nm to that at 1064 nm versus the 1064-nm power density. The 1064-nm power density is also shown in units of its Rabi frequency divided by twice the square root of the product of its detuning (6 cm^{-1}) and the Doppler linewidth (0.0813 cm^{-1}). The matrix element used to calculate this Rabi frequency is 1.20 (atomic units), as measured by Field.[8]

At a critical value of the 1064-nm power density we observe an enhancement of the generated 283-nm signal, relative to its small field value, of a factor of 59.

In Fig. 4 the 1064- and 355-nm power densities are held fixed at 21 MW/cm^2 and 130 kW/cm^2, respectively, and the 606-nm power density is varied through approximately three orders of magnitude. We observe a conversion efficiency over this range of $\sim 10^{-3}$. A similar, nominally flat conversion efficiency is also obtained if the 355-nm power density

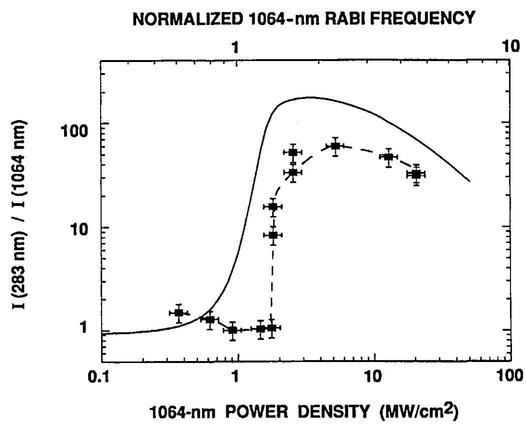

Fig. 3. Conversion efficiency (283-nm power density/ 1064-nm power density) versus 1064-nm power density, where unity on the vertical scale corresponds to an absolute conversion efficiency of 4.1×10^{-10}. The 1064-nm power density is shown in absolute units on the lower scale and, on the upper scale, in units of Rabi frequency divided by twice the square root of the product of the Doppler width and the 6-cm^{-1} detuning. The theoretical curve (solid curve) is in absolute units and is not fitted.

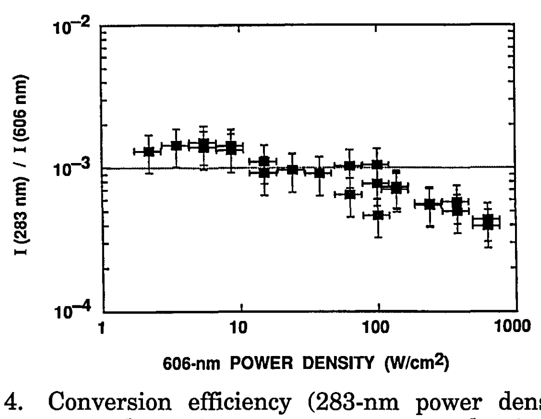

Fig. 4. Conversion efficiency (283-nm power density/ 606-nm power density) versus 606-nm power density.

is varied and the other power densities are held constant.

The solid curve in Fig. 3 shows the calculated conversion efficiency in absolute units for the parameters of this experiment. This calculation is carried out for each spectral component ω of the 606-nm signal as follows: The linear and nonlinear susceptibilities $\chi^{(1)}$ and $\chi^{(3)}$ are calculated from the formulas of Ref. 2; these susceptibilities are averaged over the Doppler linewidth to produce the macroscopic susceptibilities $\chi_D^{(1)}(\omega)$ and $\chi_D^{(3)}(\omega)$; and the generated radiation at 283 nm is calculated from Maxwell's equations. The total generated power is then obtained by integrating over all ω. The parameters for the calculated curve are 606-nm linewidth = 0.03 cm^{-1}; Doppler linewidth, 0.0813 cm^{-1}; $|1\rangle-|2\rangle$ homogeneous linewidth, 0.00070 cm^{-1}; $|1\rangle-|3\rangle$ homogeneous linewidth, 0.0098 cm^{-1}; Pb atom density, 2.5×10^{16} atoms/cm^3; cell length, 9 cm; and matrix elements $\mu_{13} = 0.808$ atomic unit and $\mu_{23} = 1.20$ atomic units.

The principal limitation of this experiment is the relatively broad linewidth (0.03 cm^{-1}) of the 606-nm laser relative to the homogeneous linewidth (0.00070 cm^{-1}) of the $|1\rangle-|2\rangle$ transition. If the 606-nm laser were monochromatic and the cell were several absorption depths long, the expected enhancement in the conversion efficiency when the inhomogeneous linewidth is overcome is equal to $1/(\pi \ln 2)$ times the square of the ratio of the inhomogeneous-to-homogeneous linewidth of the $|1\rangle-|2\rangle$ transition[2] and, for the parameters of this experiment, is a factor of 6200. For our experiment, only the portion of the 606-nm laser linewidth that is within ~0.0008 cm^{-1} is electromagnetically phase matched and converted to 283 nm. One may therefore infer a conversion efficiency for this narrow linewidth of approximately 4%. The low conversion efficiency of 2.4×10^{-8}, as referenced to the power density of the 1064-nm laser, is a result of the small available power in the phase-matchable 606-nm laser linewidth and also, to some extent, a result of the 6-cm^{-1} detuning, which causes the 1064-nm power density to be approximately ten times larger than would otherwise be required.

We have shown that there is a critical laser intensity at which the inhomogeneous atomic broadening is overcome, thereby permitting phase matching and a steplike increase in the conversion efficiency.

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