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## Third-Harmonic Generation in Phase-Matched Rb Vapor*

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We report the generation of 0.3547-$\mu$m radiation by tripling 1.064-$\mu$m radiation in a phase-matched mixture of rubidium and xenon. We find $\chi(3\omega)=1.4\times10^{-32}$ esu, which is about $10^6$ times greater than that of He. Phase matching occurs for Xe and Rb atoms in the ratio 412:1.

This Letter reports the generation of 0.3547-$\mu$m radiation by tripling 1.064-$\mu$m radiation in a phase-matched mixture of rubidium vapor and xenon.[1] We experimentally confirm a third-harmonic nonlinear susceptibility in Rb vapor which is about $10^6$ times greater than that of He.[2] This large susceptibility is due to the large oscillator strengths and resonant enhancements resulting from Rb transitions in the near infrared, visible, and uv. The closest of these transitions is 3200 $cm^{-1}$ from 1.064 $\mu$m and 315 $cm^{-1}$ from 0.3547 $\mu$m, and thus significant loss is not introduced at either of these frequencies.

As a result of the large oscillator strengths of the 0.7800- and 0.7948-$\mu$m transitions ($f_{ij}=0.9$ and 0.3, respectively), the refractive index of Rb vapor at 1.064 $\mu$m is greater than its refractive index at 0.3547 $\mu$m. The addition of Xe, in the ratio of 412 atoms of Xe to each atom of Rb, causes the refractive index at 1.064 $\mu$m to equal the refractive index at 0.3547 $\mu$m (achieves phase matching) and increases the generated third-harmonic power by $(L/L_c)^2$, where $L$ is the cell length and $L_c$ is the coherence length of the Rb vapor in the absence of Xe. In the experiments reported here we have observed a phase-matching third-harmonic power enhancement of a factor of 33, and believe that much greater enhancements will be possible with engineering improvements.

The possibility of using anomalous dispersion to achieve phase matching was suggested by a number of early nonlinear-optics workers,[3] and has been demonstrated in liquids by Bey, Giuliani, and Rabin.[4]

The experimental apparatus consisted of an acousto-optically $Q$-switched Nd-doped yttrium aluminum garnet laser and amplifier which furnished up to 100 kW of $TEM_{00}$-mode radiation at 1.064 $\mu$m. The Rb cells were 19 cm long and were constructed of Pyrex. The Rb metal was placed in a side arm which was maintained at a somewhat lower temperature than the main cell. By controlling the side-arm temperature from about 100 to 320°C, the vapor pressure of Rb could be continuously varied between $2\times10^{-4}$ and 2 Torr. Different pressures of Xe were placed in the cells before sealing. The 0.3547-$\mu$m light was detected using an RCA 1P28 photomultiplier with an S5 photocathode, which followed filters and a monochromator used to discriminate against the 1.064-$\mu$m radiation.

The first experiment was performed without Xe present and was aimed at determining the nonlinearity of the Rb vapor. The incident laser beam was focused to a beam diameter of 0.52 mm positioned at the output window of the Rb cell. The confocal parameter for this focus was 40 cm, thus yielding a slightly converging beam over the length of the Rb cell. The points in Fig. 1 show generated third-harmonic power as a function of the temperature of the Rb side arm. The solid

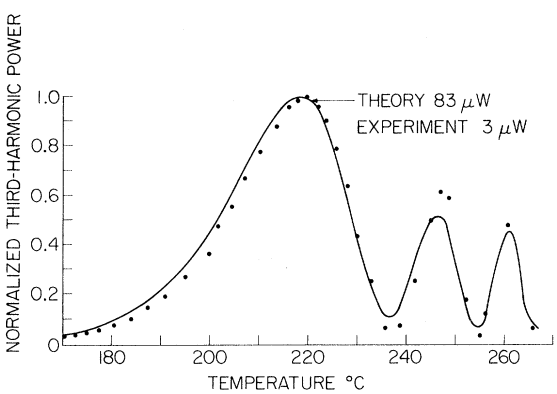


FIG. 1. Normalized third-harmonic power versus temperature for pure Rb vapor. The 50-kW 1.064-μm beam was focused on the output cell window with a confocal parameter of 40 cm.

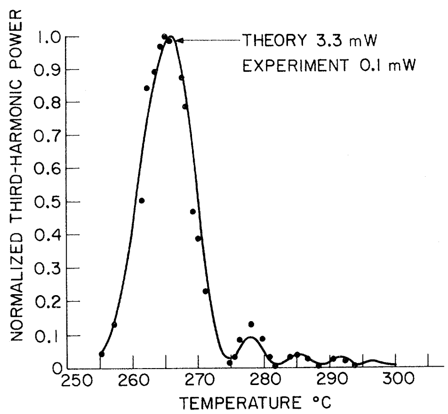


FIG. 2. Normalized third-harmonic power versus temperature for Rb with 81 Torr Xe at 20°C. Experimental points were uniformly translated toward cooler temperatures by 5°C. Incident power, focusing, and confocal parameter are as in Fig. 1.

curve is obtained from

$$\frac{P_3}{P_1^{\,3}}=\frac{3\pi^2N^2\chi^2}{\lambda^4c^2\epsilon_0^{\,4}g^2}\left|\int_{-\zeta}^{0}\frac{\exp(\frac{1}{2}jb\Delta k\xi')}{(1+j\xi')^2}\,d\xi'\right|^2 \text{ (mks)}, \qquad (1)$$

which is obtained by appropriately integrating an expression given by Ward,[2] where $P_3$ and $P_1$ are the third-harmonic and fundamental powers, respectively, $N$ is the number of Rb atoms/cm$^3$, $g=2$ is the degeneracy of the 5$s$ level, $\chi$ is the third-harmonic susceptibility ($\chi_{\rm mks}=\frac{1}{81}\times10^{-17}\chi_{\rm esu}$), $\lambda_1$ is the fundamental wavelength, and $b$ is the confocal parameter of the fundamental beam—assumed to be focused on the output window of the gas cell. $\zeta$, a normalized $z$ coordinate, is related to the position of the focus at $z=f$ by $\zeta=2z/b$. The $\vec{k}$ mismatch $\Delta k=6\pi(n_1-n_3)/\lambda_1$ is obtained from the Sellmeier equations for Rb.

For a nearly collimated beam, $b\gg L$, the right-hand side of Eq. (1) is proportional to $\sin^2(\frac{1}{2}\pi L/L_c)$, where $L_c=\pi/\Delta k$; this predicts maximum output power for a Rb vapor pressure such that $L_c$ equals the cell length $L$. Based on the Sellmeier equation for Rb, $L_c=19$ cm (our cell length) for $N=1.14\times10^5$ atoms/cm$^3$, which occurs at a temperature of $T=210$°C. As a result of the converging input beam, Eq. (1) predicts that the height of the first lobe should exceed that of the latter lobes and should occur about 9°C hotter than would be the case for a perfectly plane wave. Both experimental and theoretical results were normalized to a peak amplitude of unity. The excellent agreement of periodicities indicates that the Sellmeier equation for Rb is accurately known, and that the Rb temperature was correctly measured.

With an input power $P_1=50$ kW and a confocal parameter $b=40$ cm (thus a peak power density of about 47 MW/cm$^2$) we obtained a third-harmonic power $P_3=3$ μW at $T=219$°C. At this temperature there are $1.59\times10^{15}$ Rb atoms/cm$^3$ in the cell. From Eq. (1) we obtain $\chi_{\rm Rb}(\text{meas})=1.4\times10^{-32}$ esu. Considering experimental uncertainties, this agrees reasonably well with the previously calculated value $\chi_{\rm Rb}(\text{calc})=7.42\times10^{-32}$ esu.[1]

The second experiment was aimed at demonstrating that the Rb vapor could be phase matched by introducing a normally dispersive gas such as Xe, thus allowing the interaction to extend over many coherence lengths. In this experiment, 81 Torr of xenon at 20°C was introduced into the cell before sealing. From the Sellmeier equations of Xe and Rb we calculate that phase matching should occur at a Rb vapor pressure corresponding to a cell temperature of $T=262$°C.

Experimental results are shown in Fig. 2. Note that for the same input power and focus, the peak power obtained in the phase-matched case exceeds that of the pure-Rb case (Fig. 1) by a factor of 33. To achieve the best experimental-theoretical fit, it was necessary to translate the experimental points uniformly by 5°C. Fitting the peak experimentally observed power by the peak of the theoretical curve of Eq. (1), with the use of Sellmeier equations for both Rb and Xe, yields $\chi_{\rm Rb}=1.3\times10^{-32}$ esu, in close agreement with the pure Rb case.

In the course of the experimental work, two problems were encountered. First, Rb vapor reacts with Pyrex and leads to cell yellowing and uv opacity after several hours at temperatures above about 300°C. Second, the low Rb diffusion rate in Xe combined with the Rb reaction or clean-up problem limited the amount of Rb which was uniformly obtainable over our cell length and prevented experiments at higher Rb-Xe pressures. We believe that both of these problems can be solved by employing a heat-pipe oven of the type recently described by Vidal and Cooper.[5]

In summary, we have measured $\chi_{Rb} = 1.4\times10^{-32}$ esu for tripling 1.064 to 0.3547 $\mu$m, and have demonstrated that metal vapors may be phase matched via the addition of inert gases. Based on these measurements and on previous calculations,[1] we believe that if the Rb vapor pressure could be increased to 16 Torr (415°C) and the cell length extended to 50 cm, 50% conversion efficiency to 0.35 $\mu$m should be obtainable with an input power of about 10 MW. Peak powers exceeding this are now readily available with picosecond lasers, and calculations have shown that subpicosecond pulses are acceptable in a system of this type.[1]

Since metal vapors are often nearly transparent for wavelengths above their ionization potentials,[6] this technique should allow tripling of 6943 Å in Na and also tripling of tunable dye lasers. It should also be possible to cascade several similar systems to extend this technique through the vacuum ultraviolet. For example, Cd-He is of interest for 3547 → 1182 Å generation.

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# Computer Simulation of Anomalous dc Resistivity

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The current-driven ion-sound instability is investigated by two-dimensional computer experiments. It is found that, in contrast to the one-dimensional case, the instability produces an anomalous resistivity.

The two-stream instability excited by a strong electric current in a plasma has been the subject of numerous investigations, since it is believed to cause anomalous resistivity and turbulent heating in many plasma devices. Though a linear stability analysis is rather simple, little is known so far about the nonlinear behavior. Theoretical methods are rather limited in the case of a strongly turbulent system, while experimental observations normally do not admit of simple interpretation since various effects interfere. Here numerical simulation experiments can be very useful.

Recently, the two-stream instability driven by a constant external electric field $E_0$ has been studied by numerical simulation of a one-dimensional plasma.[1,2] The main result was that under the combined influence of the instability and the driving field the electrons are accelerated and heated in such a way that the drift velocity $v_d$ tends to be close to the thermal velocity $v_{\text{th},e}$, $v_d \simeq v_{\text{th},e}$, which means that the current increases linearly with time with half the free acceleration rate, and that there is no resistivity in the usual sense. This behavior seems to be in clear contrast to observations in many experiments, implying that the model of a one-dimensional plas-