Kavanagh, J. M. Khan, C. B. Layne, and E. J. Zaharis, Phys. Rev. Lett. 24, 931 (1970).
[8]D. L. Walters and C. P. Bhalla, Phys. Rev. A 4, 2164 (1971).
[9]R. K. Cacak, Q. C. Kessel, and M. E. Rudd, Phys. Rev. A 2, 1327 (1970).
[10]I. A. Sellin, C. D. Moak, P. M. Griffin, and J. A. Biggerstaff, Phys. Rev. 184, 56 (1969); H. J. Andrä, Phys. Rev. Lett. 25, 325 (1970); C. H. Liu, S. Bashkin, and W. S. Bickel, Phys. Rev. Lett. 26, 222 (1971); T. Hadeishi, Bull. Amer. Phys. Soc. 16, 1356 (1971).
[11]J. K. Rice and F. W. Bingham, Phys. Rev. A 5, 2134 (1972); Q. C. Kessel and E. Everhart, Phys. Rev. 146, 16 (1966).
[12]D. Powers and W. Whaling, Phys. Rev. 126, 61 (1962).
[13]Either a conventional duoplasmatron model or a more recently developed source: M. Nunogaki, J. Itoh, H. Akimune, and T. Suita, Jap. J. Appl. Phys. 10, 1640 (1971); R. C. Davis, O. B. Morgan, L. D. Stewart, and W. L. Stirling, Rev. Sci. Instrum. 43, 278 (1972).
[14]J. Lindhard, M. Scharff, and H. E. Schiøtt, Kgl. Dan. Vidensk. Selsk., Mat.-Fys. Medd. 33, No. 14 (1963); J. H. Ormrod and H. E. Duckworth, Can. J. Phys. 41, 1424 (1963); D. I. Porat and K. Ramavataram, Proc. Roy. Soc., Ser. A 252, 394 (1959); B. Fastrup, P. Hvelplund, and C. A. Saulter, Kgl. Dan. Vidensk. Selsk., Mat.-Fys. Medd. 35, No. 10 (1966).
[15]V. Ernst and P. Stehle, Phys. Rev. 176, 1456 (1968); L. Allen and G. I. Peters, Phys. Lett. 31A, 95 (1970); N. E. Rehler and J. H. Eberly, Phys. Rev. A 3, 1735 (1971); B. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A 4, 854 (1971), and earlier work referenced; F. Varsanyi, Appl. Phys. Lett. 19, 169 (1971).
[16]T. M. Kavanagh, M. E. Cunningham, R. C. Der, R. J. Fortner, J. M. Khan, E. J. Zaharis, and J. D. Garcia, Phys. Rev. Lett. 25, 1473 (1970).
[17]R. W. Fink, R. C. Jopson, H. Mark, and C. D. Swift, Rev. Mod. Phys. 38, 513 (1966); D. L. Walters and C. P. Bhalla, Phys. Rev. A 3, 1919 (1971); E. J. McGuire, Phys. Rev. A 2, 273 (1970); C. E. Dick and A. C. Lucas, Phys. Rev. A 2, 580 (1970); F. W. Saris and D. Onderdelinden, Physica (Utrecht) 49, 441 (1970).

Generation of Vacuum Ultraviolet Radiation in Phase-Matched Cd Vapor*

A. H. Kung, J. F. Young, G. C. Bjorklund, and S. E. Harris
Microwave Laboratory, Stanford University, Stanford, California 94305
(Received 17 July 1972)

We report the generation of 1773-, 1520-, and 1182-Å radiation by frequency tripling and summing in a phase-matched mixture of Cd and Ar. For the third-harmonic process, 5320→1773 Å phase matching occurs for Cd and Ar atoms in the ratio 1:25, and $\chi^{(3)} \cong 2 \times 10^{-34}$ esu/atom. The energy conversion efficiency to 1773 Å was about 10^{-4}, yielding a peak picosecond power of 7 kW.

Because of the increased difficulty of obtaining laser oscillations in the vacuum-ultraviolet (vuv) and soft x-ray regions of the electromagnetic spectrum,[1,2] it is of interest to consider other techniques which are capable of generating coherent radiation in these spectral regions. One possible approach is to use cascaded frequency triplers to convert the very high peak power now available at 1.06 μm to much shorter wavelengths. In this Letter we report the generation of 1.06-μm harmonic vuv radiation at 1773, 1520, and 1182 Å.

To obtain radiation at the above frequencies we employed the technique of phase-matched harmonic generation and frequency summing in a mixture of a metallic vapor and an inert gas. This technique was first suggested by Harris and Miles[3] and experimentally demonstrated by Young *et al.*[4] In these first experiments a mixture of Rb vapor and Xe was used for the third-harmonic process 1.06 μm → 3547 Å. To extend this technique to the vuv we employed a mixture of Cd and Ar. The choice of Cd was indicated for two reasons: First, its nonlinear susceptibility in the vuv should be enhanced by its strong atomic transitions which extend from the fundamental resonance line at 2288 Å to the beginning of the continuum at 1378 Å. Second, as is the case for the alkali metals, Cd has a relatively small absorption cross section in the spectral region just above its ionization potential ($\sigma = 7 \times 10^{-20}$ cm^2 at $\lambda = 1182$ Å).[5] To obtain efficient harmonic or sum-frequency generation it is necessary that the driving dipole polarization wave travel at the same velocity as the electromagnetic wave which it is desired to generate. This phase-matched condition is obtained by correctly choosing the ratio of Cd to Ar atoms.

We describe three nonlinear processes. These are (1) tripling of 5320 Å to yield 1773 Å, (2) summing of 1.064 μm with 3547 Å to yield 1520 Å, and (3) tripling of 3547 Å to yield 1182 Å. The exper-

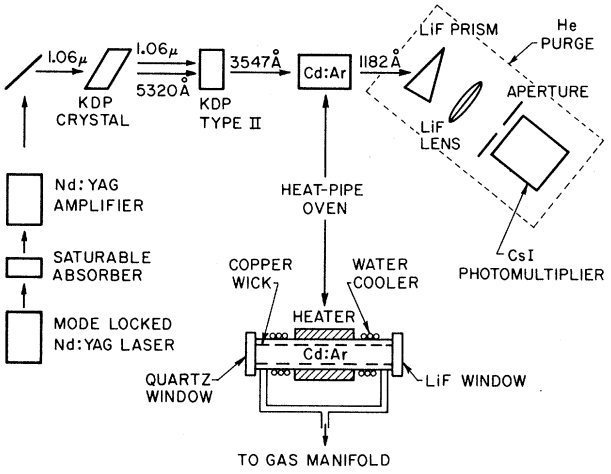

FIG. 1. Schematic of experimental apparatus for generation of 1182 Å.

imental apparatus is shown schematically in Fig. 1. As the 1.064-μm source we used a mode-locked Nd:YAlG (Nd-doped yttrium aluminum garnet) laser followed by a Kodak 9740 saturable dye absorber cell and a Nd:YAlG amplifier. The resulting 1.064-μm output consisted of about ten pulses, spaced by 5 nsec, and each about 50 psec long. The total energy of these pulses was about 10 mJ, yielding a peak power of about 20 MW. A potassium dihydrogen phosphate crystal doubled this radiation to 5320 Å with an energy conversion efficiency of up to 80%. For the experiments requiring 3547 Å, this efficiency was reduced to 50% and a second, type-II phase-matched potassium dihydrogen phosphate crystal was used to sum the remaining 1.064-μm radiation with the 5320-Å radiation to yield 3547 Å at an overall 1.064-μm to 3547-Å energy conversion of about 10%.

The Cd cell was modeled after the open-ended heat-pipe oven described by Vidal and Cooper[6] and consisted of a copper wick inside a stainless steel tube, with water cooling to protect the quartz (input) and LiF (output) end windows. The cell was wound with a 20-cm-long heating coil and had an inner diameter of 2.5 cm. Based on thermocouple measurements on the outer surface of the oven, we estimate that temperature was constant to within ± 1°C (the estimated necessary temperature tolerance) over about a 2-cm length. Ar or He was introduced near the ends of the cell and appeared to mix homogeneously with the Cd vapor in several seconds.

The generated vuv radiation was incident on a homemade, He-purged, LiF-prism monochromator, and was detected using a solar-blind EMR 542-G photomultiplier with a CsI photocathode.

A first set of experiments was aimed at determining whether the Cd vapor was negatively dispersive and thus whether it would be possible to obtain phase matching by adding a normally dispersive buffer gas. Generated third-harmonic power at 1773 Å is shown as a function of cell temperature in Fig. 2. An important feature is the fact that the height of the first lobe exceeds that of the latter lobes. This only occurs in a negatively dispersive medium, and results since in such a medium, off-axis converging and diverging light rays are in effect phase matched.[7,8] In this experiment the 5320-Å radiation is focused to a beam with a confocal parameter of $b = 100$ cm. As the cell temperature and thus the density of the Cd vapor is further increased, the cell becomes an increasing number of coherence lengths long, resulting in the oscillating output power. From the fringe spacing versus temperature, an effective cell length ($L = 2$ cm), and the vapor-pressure curve of Cd, the coherence length may be estimated. Table I gives the coherence length at 10^{17} atoms/cm^3 ($p = 8$ Torr, $T = 475$°C) for the first two of the nonlinear processes considered in this Letter. The signal at 1182 Å was at just about the noise level produced by scattered 3547-Å light, and a measurement of its coherence

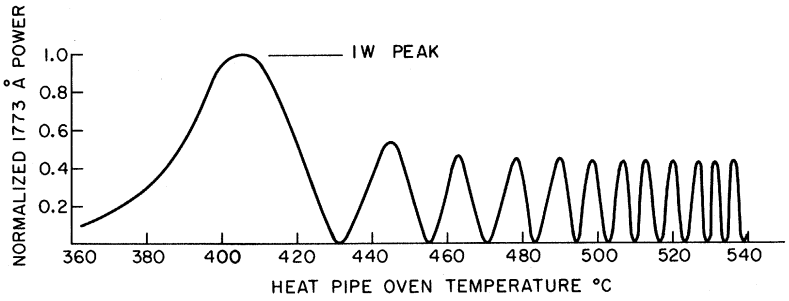

FIG. 2. Normalized 1773-Å output power versus oven temperature without phase matching. 5320-Å input power, 20 MW. Confocal parameter of input beam, 100 cm.

TABLE I. Summary of experimental data.

Input wavelengths (Å)	Generated wavelength (Å)	L_c at 10^{17} atom/cm^3 (cm)	Phase-matching ratio $N_{Cd}:N_{Ar}$	Energy conversion efficiency	$\chi^{(3)}$ (esu/atom)
5320	1773	−0.57	1:25	10^{-4}	2×10^{-34}
1.064×10^4, 3547	1520	−0.23	1:15	10^{-6}	2×10^{-33}
3547	1182	•••	1:2.5	10^{-7}	•••

length could not be made. To prevent condensation of the Cd vapor on the cell end windows, 455 Torr of He was also present in the cell; however, its dispersion is sufficiently small that the correction introduced to the measured coherence lengths is negligible.

In the next set of experiments, phase matching was obtained by establishing an appropriate Cd:Ar ratio. Experimental results for phase-matched 1773-Å generation are shown in Fig. 3. With 700 Torr of Ar present, the phase-matched peak occurred at a cell temperature of 536°C, corresponding to a Cd:Ar ratio of 1:25. For the same input power and focusing, the phase-matched peak power is 30 times greater than that obtained for the Cd:He combination. The failure of the higher temperature side lobes to decrease as rapidly as expected is probably due to temperature, and thus vapor density, gradients within the cell.

In another experiment at 1773 Å, we reduced the confocal parameter of the incident 0.53-μm beam to 5 cm, and at 700 Torr of argon obtained an energy conversion efficiency of about 10^{-4}, corresponding to a peak picosecond power of about 7 kW. Based on the effective cell length, power density, and vapor pressure of Cd, we obtain $\chi^{(3)}=1\times10^{-34}$ esu/atom. Based on a normalized measurement to the third-harmonic power generated in the last coherence length of a LiF crystal, we obtain $\chi^{(3)}=2\times10^{-34}$ esu/atom.

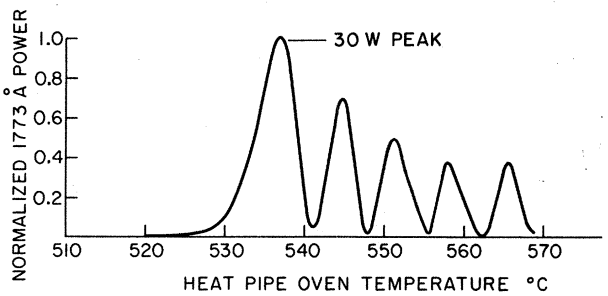

FIG. 3. Normalized 1773-Å output power versus oven temperature with 700 Torr (at 536°C) of Ar present. Input power and focus are the same as in Fig. 2. Note the enhancement of peak output power by a factor of 30.

For the generation of 1520-Å radiation, both the 3547-Å and the 1.064-μm beams were focused to a confocal parameter of 20 cm. For 100 Torr of argon, phase matching occurred at 460°C at an Ar:Cd ratio of 15:1, and the enhancement due to argon, as compared to the phase-unmatched case, was a factor of 8.5. For this process the observed energy conversion efficiency was about 10^{-6}, which yields a lower bound of $\chi^{(3)}\cong2\times10^{-33}$ esu/atom. For 1182-Å generation, phase matching to 200 Torr of Ar occurred at 579°C at an Ar:Cd ratio of 2.5:1, and the observed energy conversion was about 10^{-7}. Experimental results are summarized in Table I.

In considering the results of these experiments it is clear that conversion efficiencies should be improved if this process is to yield practical devices. Based on the measured $\chi^{(3)}\cong2\times10^{-34}$ esu/atom for 1773 Å, a 50-cm-long cell with a Cd vapor pressure of 20 Torr would yield 50% peak power conversion efficiency for an incident confocally focused beam with a peak power of about 57 MW. This assumes that the harmonic process does not saturate, which at high incident pulse energies may not be the case. The dominant saturation process is caused by small absorption of the fundamental or third-harmonic frequency by the metal vapor. As atoms are excited to higher states, the metal-vapor refractive index is reduced, the phase-matching condition broken, and the beam thermally defocused. These problems are under study in a Na:Xe system and results will be reported subsequently.[8] Other practical problems such as maintaining a zone of sufficiently constant temperature and homogeneity over the required length must also be solved.

We note that radiation generated by this technique preserves the characteristics of the lower-frequency laser radiation. It is thus diffraction limited, polarized, of picosecond time scale, and of relatively narrow bandwidth. We believe that high efficiencies will be obtained, and experiments to extend the technique further into the vuv

are underway.

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[1]R. T. Hodgson and R. W. Dreyfus, Phys. Rev. Lett. 28, 536 (1972); W. Waynant, Phys. Rev. Lett. 28, 533 (1972).

[2]M. A. Duguay and P. M. Rentzepis, Appl. Phys. Lett. 10, 350 (1967).

[3]S. E. Harris and R. B. Miles, Appl. Phys. Lett. 19, 385 (1971).

[4]J. F. Young, G. C. Bjorklund, A. H. Kung, R. B. Miles, and S. E. Harris, Phys. Rev. Lett. 27, 1551 (1971).

[5]R. B. Cairns, H. Harrison, and R. I. Schoen, J. Chem. Phys. 51, 5440 (1969).

[6]C. R. Vidal and J. Cooper, J. Appl. Phys. 40, 3370 (1969).

[7]J. F. Ward and G. H. C. New, Phys. Rev. 185, 57 (1969).

[8]R. B. Miles and S. E. Harris, "Optical Third Harmonic Generation in Alkali Metal Vapors" (to be published).

Thermodynamic Evidence Against a Quadratic Term in the Phonon Spectrum of Superfluid Helium*

C. Zasada and R. K. Pathria
Department of Physics, University of Waterloo, Waterloo, Ontario, Canada
(Received 14 August 1972)

A quadratic term, characterized by the coefficient α_1, in the phonon spectrum $\epsilon(k) = c\hbar k(1-\alpha_1 k-\alpha_2 k^2-\cdots)$ is shown to be inconsistent with data on the low-temperature specific heat of liquid He II. Consistency obtains if α_1 is set identically equal to zero.

The continuing controversy over the analytic form of the low-momentum excitation spectrum of liquid He II was initiated by the suggestion of Maris and Massey[1] that the coefficient γ in the expression

$$\epsilon(p) = cp(1-\gamma p^2-\delta p^4-\cdots), \quad (1)$$

which was generally believed to be positive, may in fact be negative. This would allow a reconciliation of the experimental data on the attenuation and velocity of sound in liquid He II with the current theories of superfluidity. Further evidence for negative γ has come from the analysis of low-temperature specific-heat data[2] and from x-ray scattering measurements.[3] In the meantime, Feenberg[4] has shown that in the case of a dilute, weakly interacting Bose gas, if the interatomic potential falls off asymptotically as $(1/r)^6$, then the energy spectrum of elementary excitations may contain both odd and even powers of p—with the exception of a term in p^2. On the other hand, Molinari and Regge,[5] who analyzed the neutron scattering data of Woods and Cowley[6] to test various analytic properties of the phonon dispersion curve, have concluded that a p^2 term may as well be present. So, quite generally, one may write

$$\epsilon(k) = c\hbar k(1-\alpha_1 k-\alpha_2 k^2-\cdots). \quad (2)$$

The conclusion of Molinari and Regge has been supported by the recent work of Anderson and Sabisky,[7] who have studied dispersion in the phase velocity of first sound in superfluid helium at 1.38°K in the frequency range 20–60 GHz; their value of α_1 agrees favorably with the one obtained by Molinari and Regge, viz., 0.27 Å. Most recently, however, Roach *et al.*[8] have carried out a direct measurement of the velocity of 30- and 90-MHz sound waves in helium below 0.1°K and have shown that the value of α_1 cannot be larger than 0.01 Å. Accordingly, the controversy has now shifted from the cubic to the quadratic term in the spectrum.

To help resolve this controversy we undertook a re-examination of the specific-heat data of Phillips, Waterfield, and Hoffer[2] by including a nonzero quadratic term in the phonon spectrum and comparing the resulting values of the various parameters of the excitation spectrum with the values obtaining from other, more direct, sources (such as Abraham *et al.*[9] and Donnelly[10]). In particular, we studied the density dependence of these parameters. It turns out that the inclusion of a quadratic term in the phonon spectrum leads to a set of parameters whose behavior, as a function of density, is too erratic to be acceptable.