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TUNABLE COHERENT PARAMETRIC OSCILLATION IN $LiNbO_3$ AT OPTICAL FREQUENCIES

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We wish to report the observation of coherent, tunable, optical parametric oscillation. This has been observed in lithium metaniobate in the wavelength range 0.97 to 1.15 μ, with optical pumping at wavelength 0.529 μ. The results of our experiments suggest that oscillators of this type may be continuously tuned through a significant fraction of the visible and near infrared spectrum. Discussions of optical parametric amplifiers and oscillators have been given by Kingston,[1] Kroll,[2] Akhmanov and Khokhlov,[3] Armstrong et al.,[4] and other authors. A general treatment of the parametric amplification principle has been given by Louisell.[5] Recent observations of difference frequency generation in KH_2PO_4 by Wang and Racette[6] have indicated significant parametric amplification. Ashkin and Boyd[7] have observed difference frequency generation in $LiNbO_3$ using two gas lasers.

The experimental arrangement is shown in Fig. 1. Central to the experiment are two high optical quality crystals of ferroelectric $LiNbO_3$.[8] The nonlinear optical properties of $LiNbO_3$ have been reported earlier.[9,10] These crystals were grown by the Czochralski technique[11,12] as single-domain samples[13] using methods developed by Nassau.[8,12] The pump light at 0.529 μ, polarized normal to the diagram, was obtained by harmonic generation in crystal No. 1 from a conventional $CaWO_4$:Nd^{+3} rotating-prism giant-pulse laser.[14] The harmonic generator crystal was 0.61 cm thick with plane parallel uncoated optical surfaces normal to the beam; its optic axis was in the plane of its surfaces to ±0.15°, and oriented very nearly normal to the plane of the figure.

The oscillator crystal, No. 2, 0.539 cm long and 0.5 cm in diameter, was coated with dielec-

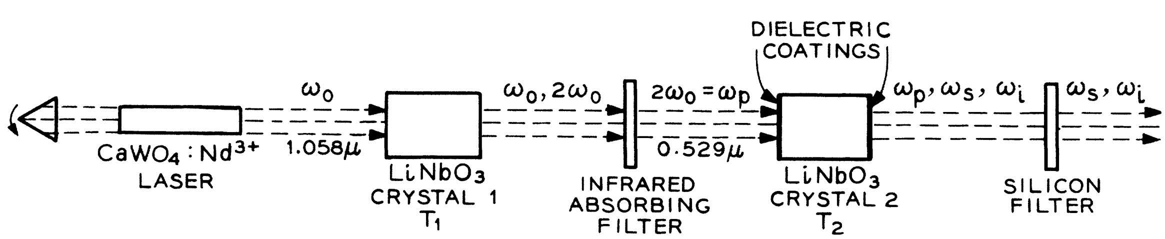

FIG. 1. Optical parametric oscillator apparatus. ω_p, ω_s, and ω_i are the pump, signal, and idler frequencies, respectively.

tric films to form an optical resonator. The crystal was cut with the optic axis in the plane of the crystal face to within $\frac{3}{4}$ deg and oriented approximately normal to the diagram. The entrance and exit surfaces were polished plane parallel to ±1 sec. The dielectric coatings were designed for peak reflectivity at 1.058 μ; the transmission of each coating was less than 0.4% throughout the range 0.96 to 1.16 μ. The transmission was 50% at 0.529 μ. The effective cavity reflectivity was measured by observation of the angular dependence of the resonator transmission in a He-Ne laser beam at 1.15 μ. The measured finesse[15] $\mathfrak{F}$ of 14.5 at room temperature indicated an effective reflectivity R of approximately 0.80, with scattering and other internal crystal losses included in $1-R$.

We denote the beam direction and the outgoing normal to the diagram shown in Fig. 1 with the unit vectors $\hat{x}$ and $\hat{z}$, respectively. Then the extraordinary pump field $E_p\hat{z}$ is coupled to ordinary signal and idler fields $E_s\hat{y}$ and $E_i\hat{y}$ through the nonlinear polarization components, $P_z = d_{31}E_y^2$ and $P_y = 2d_{15}E_yE_z$. The nonlinear coefficient $d_{31} = 3.6\times10^{-8}$ esu, and by general symmetry conditions[4,16] is equal to d_{15}.

We assume that the signal, idler, and pump waves are all plane and collinear. The pump power requirements for oscillation can be seen as follows. In coupled wave notation, it is convenient to express E_p as $\frac{1}{2}\{F_p(x)\exp[i(k_px-\omega_pt)] + \text{c.c.}\}$, with similar notation for E_s and E_i.

The frequencies of the signal, idler, and pump are related by $\omega_s+\omega_i=\omega_p$; we shall further assume that the three waves are phase matched,[17] i.e.,

$$k_s+k_i=k_p. \tag{1}$$

Solution of the coupled wave equations under the condition of undepleted pump power shows a growth of the signal and idler waves of the form $F_{s,i}=F_{s0,i0}\exp(\gamma_0x)$ where the gain γ_0 is given by

$$\gamma_0^2=\frac{16\pi^4 d_{31}^2F_pF_p^*}{n_s^0n_i^0\lambda_s\lambda_i}. \tag{2}$$

In the presence of a mismatch $k_s+k_i-k_p\equiv\Delta k$, it is shown in reference 5 that the gain is given by $\gamma^2=\gamma_0^2-(\Delta k/2)^2$; the net gain disappears for $\Delta k \geq 2\gamma_0$. In Eq. (2), n_s^0 and n_i^0 represent the ordinary refractive indices, and λ_s and λ_i the free-space wavelengths, at the signal and idler frequencies, respectively. Let W represent the pump intensity in watts cm^{-2}. For $\lambda_s=\lambda_i = 1.058\times10^{-4}$ cm, $n_s^0=n_i^0=n_p^e\times2.23^{19}$, one obtains a power gain $2\gamma_0=7.38\times10^{-4}\sqrt{W}$. For a crystal of length l and effective surface reflectivity $R\approx1$, the oscillation condition is given approximately by $(\gamma_0 l)=1-R$. For $R=0.80$ the required value of γ_0 is 0.37 cm^{-1} and corresponds to a unidirectional plane-wave pump power of 1.0×10^6 W cm^{-2}. The validity of the plane-wave approximation requires that the pump beam have an angular width small compared to $\Delta\theta_c\equiv(2n_p\lambda_p/l\mathfrak{F})^{1/2}$, the angular aperture of the central Fabry-Perot peak. For our oscillator crystal No. 2, $\Delta\theta=5.5\times10^{-3}$ rad. It should be emphasized that for propagation normal to the optic axis, the effects of the angular dependence of refractive index are negligible.[10]

The frequencies of the signal and idler are determined by the phase-matching condition (1). It has been shown that phase matching in lithium niobate can be achieved for a range of wavelengths by variation of the crystal temperature.[10] Let T_0 be the temperature for which the crystal is phase matched for collinear subharmonic generation for propagation normal to the optic axis, $\omega_p\to\omega_p/2=\omega_0$. For our oscillator, $T_0\approx49.25$°C. When $\omega_s\approx\omega_i\approx\omega_0$, the dispersion and approximately linear temperature dependence of refractive index lead to

$$\omega_s-\omega_0=\omega_0-\omega_i=\eta(T-T_0)^{1/2}. \tag{3}$$

From optical data,[9,10] it is estimated that $\eta = 4.61\times10^{13}\ \text{sec}^{-1}\ (°\text{C})^{-1/2}$, or in terms of wavelength, 275 Å $(°\text{C})^{-1/2}$.

In our experiments the resonator for the giant pulse $CaWO_4$:Nd^{+3} laser was 53 cm in length and consisted of the rotating prism, the laser rod, and crystal No. 1. The harmonic pump output of crystal No. 1 occurred in several multiple pulses of length 15 to 40 nsec. The average peak pump power for which oscillation was observed was 6.7 kW, measured after transmission through crystal No. 2. The full angular width of the pump beam was between 2 and 3 $\times10^{-3}$ rad, significantly less than $\Delta\theta_c$. The beam diameter at crystal No. 2 was approximately 2 mm. If we assume ideal resonance of the pump power in crystal No. 2, the average pump intensity inside the resonator at oscillation was 4×10^5 W cm^{-2}. The spectrum of the pump was broad and complex, having a (photographed) spectral width of 1.5 Å and consisting of 10 to 20 resolvable lines. The harmonic-generator crystal was maintained at a fixed temperature for optimum harmonic generation; the conversion efficiency for the harmonic generation was about 5%. The laser power was removed from the beam by three Schott KG-3 filters; a silicon filter beyond crystal No. 2 removed pump power from the output of the oscillator crystal. The crystals were mounted in massive silver blocks temperature-controlled to ±0.02°C.

Parametric oscillation was observed to occur in the oscillator crystal No. 2 by a photomultiplier monitoring a small fraction of the output. The oscillator output had the following characteristics. (1) A sharp well-defined threshold: Usually oscillation occurred on only one of the several pump pulses, the photomultiplier signal rising by $\sim 10^6$ from the background level at oscillation. (2) Highly collimated emission: The far-field pattern of the signal beam at 9840 Å photographed with an image converter tube had a full beam width of less than 3×10^{-3} rad, (3) High monochromaticity: The signal spectrum at 9840 Å, photographed with an image converter at the focal plane of a high-resolution grating spectrograph, consisted of a single component of width ≤ 0.3 Å or several such components separated by 0.4 Å, the longitudinal mode spacing of crystal No. 2. (4) Tunability of the signal frequency over a range of 700 Å: The spectrum was photographed at low dispersion with the aid of the image converter as a function of the temperature of crystal No. 2. The results are shown in Fig. 2. The solid line represents the best fit to the data with Eq. (3) and indicates a value of η of 4.90×10^{13} $\mathrm{sec}^{-1}\,(^\circ\mathrm{C})^{-1/2}$. The representative idler wavelengths shown were observed by means of photomultiplier and slit arrangements. Oscillations could not be observed at wavelengths closer than about 200 Å to the subharmonic frequency, perhaps because of the increasing sensitivity of the k-matching condition to small temperature gradients. The output power at 9840 Å, corrected for filter absorption, was approximately 15 W. Since the coating transmission of our crystal can be increased by more than an order of magnitude without competing with other losses, it appears that the parametric oscillator should be capable of much higher efficiency. The polarization of the signal output was verified to be ordinary as expected.

The attainment of threshold apparently depends strongly on the details of the mode structure of the pump. Parametric oscillation occurred for only about one laser flash in five having the required peak power; on individual laser flashes, oscillation frequently occurred on an individual pulse other than the most intense. Since oscillation requires simultaneous cavity resonance at the signal and idler frequencies, and therefore at the pump frequency,[18] reliable oscillation will require locking the pump to resonant frequencies of crystal No. 2.

We believe that the technique described in this Letter can be extended with larger crystals and perhaps other pump sources to produce an oscillator continuously tunable over a large fraction of the visible and near infrared spectrum. With single-mode gas laser sources and focused beam techniques, this may also be possible on a cw basis.[7] It should be noted that a transverse electric field applied to the oscillator crystal can be used to modulate or stabilize the output frequency rapidly and sensitively through the linear electro-optic effect.

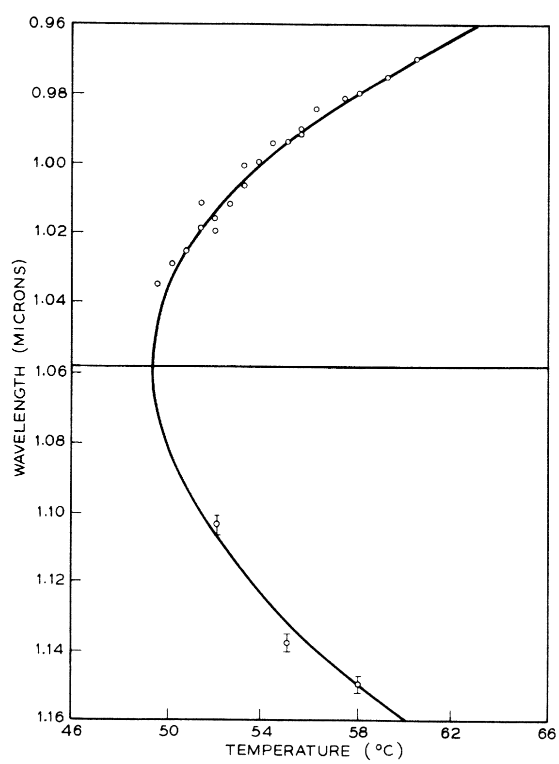

FIG. 2. Signal and idler wavelength as a function of temperature T_2 of the oscillator crystal No. 2.

We are greatly indebted to Dr. K. Nassau who grew and generously provided the unusually fine quality $LiNbO_3$ crystals, and to Dr. R. L. Batdorf and the polishing shop for fabricating them into the required flat and parallel geometry. During the course of the research, we have benefited from numerous stimulating and informative discussions with Dr. A. Ashkin and Dr. G. D. Boyd. Finally, we would like to acknowledge the patient and skillful technical assistance of Mr. A. Savage and Mr. K. Wecht.

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QUANTIZED VORTICES AND THE SUPERFLUID HELIUM ANALOG OF THE ac JOSEPHSON EFFECT

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In a recent paper Richards and Anderson have reported the observation of an effect in superfluid helium which they have interpreted in close analogy to the ac Josephson effect in superconductors.[1] Their interpretation is based on an equation which has been derived using the concept of a complex order parameter for the liquid. It is the purpose of this note to point out that this equation can also be derived from the conventional equations of two-fluid hydrodynamics combined with the assumption of quantization of vorticity in the superfluid. Although the hydrodynamic approach makes no reference to an order parameter, it is believed that this approach is closely related to the order-parameter approach and that the two points of view complement one another.

Let us consider the situation shown in Fig. 1, in which two reservoirs of superfluid helium are connected by a small circular orifice of radius b. Richards and Anderson point out that if there exists a difference $\Delta\mu$ between the chemical potentials of the two baths, the relative phase of the order parameters of the baths will vary with time at a mean rate $\omega = \Delta\mu/\hbar$. Thus, if the two baths are at the same temperature and if there is a difference of head Δz between them, the relation $\omega = mg\Delta z/\hbar$ will hold. Here m is the mass of the helium atom and g the acceleration of gravity. Richards

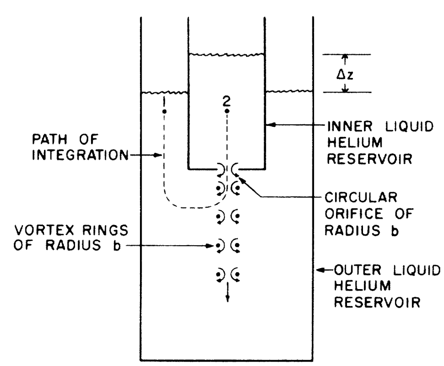

FIG. 1. Sketch illustrating the possible steady-state production of vortex rings as superfluid helium flows from the inner to the outer reservoir.