ter's analysis can be extended to finite temperatures.

[6]Ref. 4, footnote 14a.

[7]See, for example, A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinski, Quantum Field Theory in Statistical Physics (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963), p. 328.

[8]It is assumed that there is no scattering within a given grain, and the electronic energy spectrum of pure Al can be represented by that of free electrons with an effective mass m.

[9]See Ref. 7, pp. 329-341.

[10]It can be shown that terms of order $E_F(p_F l_{eff})^{-2}$ give rise to an increased density of electronic states $N(0)$. Such an effect could result in an enhanced T_c.

[11]See, for example, P. G. de Gennes, Superconductivity in Metals and Alloys (W. A. Benjamin, Inc., New York, 1966), pp. 217-227.

[12]K. Maki, Physics 1, 127 (1964).

[13]J. L. Olson, Electron Transport in Metals (Interscience Publishers, Inc., New York, 1962), p. 84.

[14]Ref. 11, p. 24.

[15]O. F. Kammerer and Myron Strongin, in Proceedings of the International Symposium on Basic Problems in Thin Film-Physics, Clausthal-Göttingen, 1965 (Vandenhoeck and Ruprecht, Göttingen, Germany, 1966), p. 511.

[16]J. J. Hauser and H. C. Theuerer, Phys. Rev. 134, A198 (1964).

[17]P. G. de Gennes and M. Tinkham, Physics 1, 107 (1964). Using these authors' Eq. (IV-36), we estimate a slope of approximately −120 kG/°K, nearly an order of magnitude greater than our largest observed values.

[18]C. J. Thomson and J. M. Blatt, Phys. Letters 5, 6 (1963), considered the effect of quantization of electronic motion in one dimension. In Ref. 3, it was noted that quantization of motion in three dimensions could account for the observed enhancement. More recently, R. H. Parmenter (private communication) and E. A. Shapoval, Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt. 5, 70 (1967) [translation: JETP Letters 5, 57 (1967)], have also calculated the enhancement of T_c due to quantization of motion in small systems.

STUDY IN AMMONIUM DIHYDROGEN PHOSPHATE OF SPONTANEOUS PARAMETRIC INTERACTION TUNABLE FROM 4400 TO 16 000 Å

Douglas Magde and Herbert Mahr
Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York
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This Letter reports the quantitative experimental study of tunable parametric amplification of quantum noise in ammonium dihydrogen phosphate (ADP). Because of phase-matching conditions the parametric noise output is limited to narrow spectral regions which could be tuned from 4400 to 16 000 Å by simply rotating the ADP crystal around the pump-beam direction. A complete study of the physical parameters which determine this spontaneous parametric interaction was made and the results compared with theory. The results are also of practical interest (a) because the observed small levels of output represent the noise level of parametric light amplifiers and (b) because the measured gain allows predictions about the construction of tunable coherent light oscillators in the visible.

The extension of tunable parametric output through the visible and the direct measurement of various physical parameters of single-passage amplification without the use of external mirrors differentiates the reported measurements from previous observations of tunable parametric oscillators. Powerful coherent output in the infrared and far red using an optical cavity and a pump at 5300 Å has been observed by Giordmaine and Miller[1] and Miller and Nordland[2] in $LiNbO_3$ and by Akhmanov et al.[3] in potassium dihydrogen phosphate.

In the present arrangement the pump beam at 3472 Å, consisting of 1-MW pulses of 20-nsec duration, traversed the ADP crystal at near-normal incidence. After attenuation of the pump light by appropriate filters, parametric signal and idler radiation was detected at a distance of about 2 m by a monochromator-photomultiplier system. The presence of signal or idler output was established by an increase of signal over background radiation (mainly due to luminescence from the cutoff filters) as the crystal was rotated through the phase-matching orientation for a preselected wavelength. From the position of these peaks the tuning curve of Fig. 1 was obtained. The tuning range of 4400-16 000 Å is limited by the onset of optical absorption by ADP in the infrared. The spectral sensitivity of the photomultiplier confined the observable part of the tuning range to 4400-10 000 Å. The dashed curve of Fig. 1 shows a quadratic dependence of the change in crystal orientation $\Delta\theta$ on the

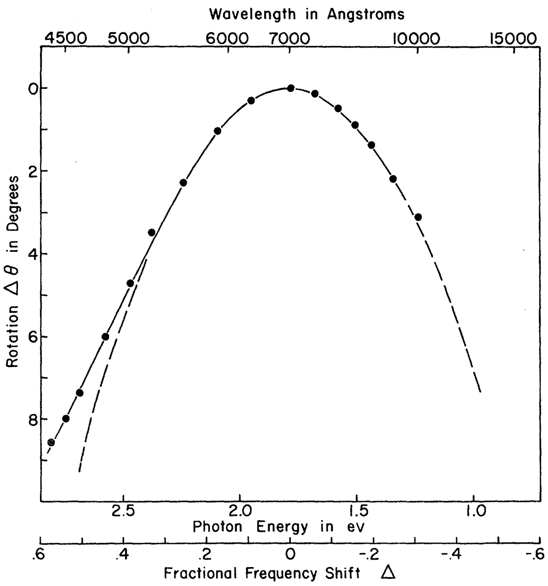

FIG. 1. The change in crystal orientation $\Delta\theta$ is plotted versus the fractional frequency shift Δ of signal and idler radiation. Δ is defined as $\Delta=(\nu-\nu_d)/\nu_d$, the shift of signal or idler frequencies from the degenerate frequency $\nu_d=\frac{1}{2}\nu_{\text{pump}}$. The dashed curve shows a quadratic dependence of $\Delta\theta$ on Δ, based on approximations to the dispersion curve of ADP.

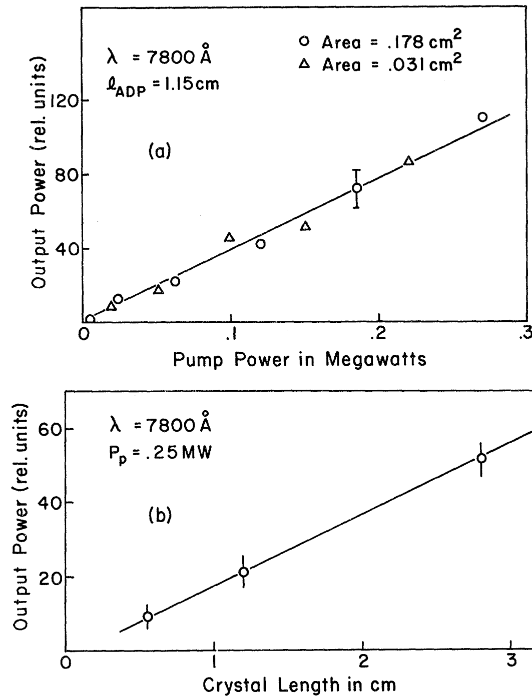

FIG. 2. (a) The observed output power is shown as a function of pump power for two different beam cross sections. (b) The output power is plotted versus the length of ADP crystals.

fractional frequency shift Δ of idler or signal radiation. This dependence is based on approximations to the dispersion curve of ADP.[4]

The effect observed is due to the spontaneous breakup, in a medium of nonlinear dielectric constant, of a pump photon $h\nu_p$ into two photons $h\nu_s$ and $h\nu_i$, such that $h\nu_p=h\nu_s+h\nu_i$.[5] Momentum conservation, or phase matching, selects that particular collinear pair of signal and idler waves for which $k_s+k_i=k_p$ in a given crystal direction. The parametric breakup arises from the presence of zero-point electromagnetic energy of one photon per mode and its output power is given by[6,7]

$$P=(\text{flux of modes which can be parametrically amplified})\times(\text{zero-point energy per mode})\times(\text{parametric gain}). \quad (1)$$

For the particular conditions of our experiment Eq. (1) becomes[7]

$$P=(n/2c)\tilde{g}^2h\nu^2[\nu^2+(d\nu/d\theta)^2]^{1/2}\Delta\Omega_p P_p l_{\text{ADP}} \quad (2)$$

with[8]

$$\tilde{g}^2=(2/\epsilon_0)(2\pi\nu_s/n_s)(2\pi\nu_i/n_i)(1/n_pc^3)\chi_2{}^2\sin^2\theta, \quad (3)$$

where ν, ν_i, and ν_s are the signal and idler frequencies; n, n_i, and n_s the indices of refraction; P_p the pump power; l_{ADP} the crystal length; $\Delta\Omega_p$ the solid angle extended by the pump beam; $d\nu/d\theta$ the inverse slope of the tuning curve at the signal or idler frequency; χ_2 the nonlinear susceptibility of ADP; and θ the phase-matching angle. Equation (2) was derived for small parametric gain ($G<1$); a coherence length $l_{\text{coh}}<l_{\text{ADP}}$; a small bandwidth of the pump beam $[\Delta\nu_p\ll(cl_{\text{coh}}/2n)]$[7,9]; and collinear interaction only.[10]

Figure 2(a) shows the observed linear dependence of the parametric output on pump power and the independence of the output on pump intensity in agreement with Eq. (2). Figure 2(b) shows a linear dependence of the output on the length of the ADP crystal. This linearity is expected from Eq. (2) for l_{ADP} larger than the coherence length. The coherence length was estimated to be about 0.3 cm from the known angular divergence of the beam and the birefringence of ADP. Figure 3 shows the measured dependence of the output on signal and idler frequencies. The full line is proportional to that term of Eq. (2), $\nu^2(d\nu/d\theta)$, which dominates

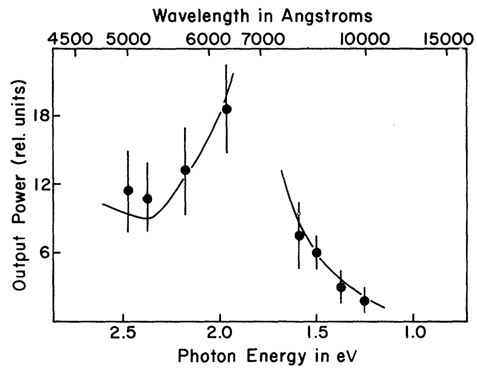

FIG. 3. The measured parametric output power is shown for various values of signal and idler frequencies ($l_{ADP}=1.15$ cm; $P_p=0.25$ MW). The solid line is proportional to $\nu^2(d\nu/d\theta)$.

the ν dependence of the output.[11] The strong dependence of the output on the slope of the tuning curve $(d\nu/d\theta)$ is a consequence of the change of the bandwidth of the emitted radiation over the tuning range. This change in bandwidth is easily measured; it agrees with the slope of the tuning curve and its value serves as an experimental determination of $\Delta\Omega_p$, the pump-beam divergence.[12]

A comparison was made at 7800 Å of the measured output with the value calculated from Eq. (2). The output power was calculated using the following values: $P_p = 2.5\times10^5$ W; $\Delta\Omega_p = 2.5\times10^{-5}$ sr; $\nu = 3.85\times10^{14}$ sec^{-1}; $n\approx 1.5$; $l_{ADP} = 1.15$ cm; $d\nu/d\theta = 3.3\times10^{15}$ rad^{-1} sec^{-1}; $\tilde{g}^2 = 3.7\times10^{-9}$ W^{-1} using[8] $\chi_2 = 6\times10^{-13}$ m/V and $\theta\approx 49.5°$. The calculated value is $P_{calc} = 0.2$ μW compared with $P_{expt} = 1$ μW.[12]

The parametric gain G is in general given by[6] $G = \sinh^2(\tilde{g}I_p^{1/2}l_{coh})$ for $l_{coh} < l_{ADP}$. For the present experimental conditions with $I_p = 4$ MW/cm^2 and $l_{coh}\approx 0.3$ cm, we calculate a gain of about 10^{-3}. It is within reason to consider an increase to $l_{coh} = 3$ cm and $I_p = 40$ MW/cm^2 with a better pump source to get exponential gain and coherent output. It might thus be possible to construct a tunable, coherent, single-passage light source in the visible by improving the beam divergence and intensity of the primary laser pump. This source would be extremely simple and would not require external mirrors for its operation.

The results demonstrate that the spontaneous parametric effect is a clear manifestation of the zero-point energy of the electromagnetic field. Experimental observation of the effect can serve as a simple tool for exploring parametric interaction.

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[1]J. A. Giordmaine and R. C. Miller, Phys. Rev. Letters 14, 973 (1965); in Physics of Quantum Electronics, edited by P. L. Kelly, B. Lax, and P. E. Tannenwald (McGraw Hill Book Company, New York, 1966), pp. 31-42; Appl. Phys. Letters 9, 293 (1966).

[2]R. C. Miller and W. A. Nordland, Appl. Phys. Letters 10, 53 (1967).

[3]S. A. Akhmanov, A. I. Kovrigin, V. A. Kolosov, A. S. Piskarskas, V. V. Fadeev, and R. V. Khokhlov, Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt. 3, 372 (1966) [translation: JETP Letters 3, 241 (1966)].

[4]A. Yariv and W. H. Louisell, IEEE J. Quantum Electron. QE-2, 418 (1966).

[5]The breakup of a pump photon can also be stimulated by the presence of signal and idler radiation. This is equivalent to large parametric gain ($G > 1$). Powerful, coherent output results in that case.

[6]W. H. Louisell, A. Yariv, and A. E. Siegman, Phys. Rev. 124, 1646 (1961).

[7]Reference 6 calculates the parametric gain per mode of noise; the importance of the factor containing the number of modes participating in the parametric interaction was pointed out to us by Professor C. L. Tang of Cornell University. There are three different physical processes which determine the size of phase space giving rise to parametric interaction: (1) allowance for phase mismatch, $\Delta k l_{coh} = \pi$; (2) allowance for noncollinear phase matching (see Ref. 10); and (3) the frequency spread of the pump $\Delta\nu_p$ or $\Delta k_p = 2\pi n\Delta\nu_p/c$. For the present experimental conditions we feel that (1), used to derive Eq. (2), gives the largest contribution.

[8]R. W. Minck, R. W. Terhune, and C. C. Wang, Appl. Opt. 5, 1595 (1966).

[9]$\Delta\nu_p$ was measured to be smaller than 3×10^9 sec^{-1}.

[10]It was pointed out to us by Professor R. V. Khokhlov of Moscow State University that noncollinear interaction will also contribute to the parametric output. For the present experimental conditions the contribution to the output from noncollinear interaction is estimated to be an order of magnitude smaller than the contribution from the collinear interaction.

[11]For the present experimental conditions, $d\nu/d\theta \gg \nu$ and, therefore, dominates in the term $[\nu^2+(d\nu/d\theta)^2]^{1/2}$ of Eq. (2). In addition, $\tilde{g}(\nu)$ depends very little on ν.

[12]Values of $\Delta\Omega_p$ obtained from the measured bandwidth of signal radiation are consistently larger by a factor of 3-4 than the values obtained with idler radiation although no such discrepancy exists between signal or idler output shown in Fig. 3. We do not understand this discrepancy and have used the lower value of $\Delta\Omega_p$ for the calculation of the output power of 0.2 μW at 7800 Å.