features.

Another consequence arises from the picture developed here. As mentioned above, the OSO-III count rate from the galactic disk exceeds the theoretical predictions quoted above by a factor that may be as large as five.[6,7] These theoretical predictions are arrived at by multiplication of the average cosmic-ray source strength with the columnar hydrogen density along the line of sight. Although the hydrogen density is known to show large fluctuations[15] this procedure was thought to be justified due to a uniform cosmic-ray density.

In the picture used here, however, the cosmic rays will stay close to their source of production within distances of the order of 100 pc for periods of 10^4-10^6 yr, depending on their speed of expansion. Inside this volume the cosmic-ray energy density will be considerably enhanced. If these sources should be correlated with regions of large hydrogen density, the usual theoretical prediction by multiplication of average values gives an answer that may easily be too low by a factor of 5.[16]

It is interesting to note that such a correlation may indeed exist. This could be so on general grounds because the sources are believed to be population-I objects. Furthermore, it has been claimed from observations that pulsars[17] and supernova remnants[18] are correlated with the spiral arms. We thus find that these correlated fluctuations of cosmic rays and gas are a very likely explanation of the high OSO-III count rate.

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[1]S. Colgate and R. H. White, Astrophys. J. 143, 626 (1966).

[2]J. P. Ostricker and J. E. Gunn, Astrophys. J. 157, 1395 (1969).

[3]R. E. Lingenfelter, Nature 224, 1182 (1969).

[4]R. Ramaty, D. V. Reames, and R. E. Lingenfelter, Phys. Rev. Lett. 24, 913 (1970).

[5]G. W. Clark, G. P. Garmire, and W. L. Kraushaar, Astrophys. J. 153, 203(L) (1970).

[6]G. W. Clark, G. P. Garmire, and W. L. Kraushaar, Proceedings of the International Astronomical Union Symposium, Rome, Italy, 1969 (to be published).

[7]G. P. Garmire, Bull. Amer. Phys. Soc. 15, 564 (1970).

[8]G. M. Frye and C. P. Wang, Astrophys. J. 158, 925 (1969).

[9]C. E. Fichtel, D. A. Kniffen, and H. B. Ögelman, Astrophys. J. 158, 193 (1969).

[10]R. Cowsik, and Y. Pal, Phys. Rev. Lett. 23, 1467 (1969).

[11]H. B. Ögelman, Nature 221, 754 (1969).

[12]J. B. Pollack and G. G. Fazio, Phys. Rev. 131, 2684 (1963).

[13]J. R. Jokipii and E. N. Parker, Astrophys. J. 155, 799 (1969).

[14]I. S. Shklovsky, *Supernovae* (Interscience, New York, (1968), Vol. XXI.

[15]A. N Bunner, P. C. Coleman, W. L. Kraushaar, D. McCammon, T. M. Palmieri, A. Shilepsky, and M. Ulmer, Proceedings of the International Astronomical Union Symposium, Rome, Italy, 1969 (to be published).

[16]This argument is due to joint discussions between J. R. Jokipii and the author.

[17]B. Y. Mills, Nature 224, 504 (1969).

[18]D. K. Milne, to be published.

COUPLED PARAMETRIC DOWNCONVERSION AND UPCONVERSION WITH SIMULTANEOUS PHASE MATCHING

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A two-step nonlinear optical interaction consisting of the spontaneous parametric downconversion and upconversion processes with simultaneous phase matching has been observed in ammonium dihydrogen phosphate. The spontaneous emission associated with upconversion in nonlinear optics is measured for the first time and found to be in reasonable agreement with theoretical estimates.

A two-step nonlinear optical interaction has been observed involving the spontaneous downconversion process,

$$\omega_p{}^e \rightarrow \omega_1{}^o + \omega_2{}^e, \qquad (1)$$

and upconversion process,

$$\omega_p{}^o + \omega_1{}^o \rightarrow \omega_3{}^e, \qquad (2)$$

under conditions in which both processes are simultaneous collinear phase matching (SCPM) of nonlinear optical processes, a special case of the spontaneous emission inherent in the upconversion process has been observed and measured for the first time.

The spontaneous downconversion process (1) has been discussed[1,2] as the noise source in the parametric upconversion process (2), where ω_1^o is common to both processes. In general, upconversion is carried out under phase-matched conditions while the noise process is greatly mismatched. For example, a 2-MW/cm^2 pump intensity in a crystal of $LiNbO_3$ produces an upconverted signal with roughly 1% conversion efficiency.[3] In this case an estimated noise of less than 10^{-14} W/cm^2 at ω_3 is anticipated[1] but has not been observed. The difficulty of measuring the noise is not only a problem of its small intensity, but also that there may be other spurious noise contributions of unknown origin which enter into the problem.[3] Hence, to verify the presence of a two-step interaction of the type described by processes (1) and (2) both must be observed under reasonably efficient conditions. The possibility of simultaneous phase matching provides this opportunity.

An analysis of a substantial number of the available nonlinear crystals and potential pump wavelengths has revealed several possibilities for observing SCPM of processes (1) and (2). These include two cases in ammonium dihydrogen phosphate (ADP) with 0.530-μm pump radiation, and with a common wavelength or ordinary polarization of either 0.874 or 1.70 μm. Other examples are in $LiNbO_3$ with a 1.06-μm pump; the common wavelengths for two cases are 1.55 and 3.15 μm, both again with ordinary polarization.

This Letter reports a detailed investigation in the case of ADP pumped with doubled neodymium at ω_p corresponding to 0.530 μm with a common wavelength λ_1^o of 0.874 μm.[4] The indicated e (extraordinary) and o (ordinary) polarizations for the waves in processes (1) and (2) are appropriate for the ADP symmetry. The tuning curves for collinear phase-matched downconversion with an extraordinary pump and collinear phase-matched upconversion with an ordinary pump are shown in Fig. 1(a). In the figure the common wavelength in the two processes λ_1^o and the upconverted wavelength λ_3^e are plotted as a function of θ, the angle between the c axis of the crystal and the direction of pump propagation. SCPM is seen to occur for $\lambda_1^o = 0.874$ μm and $\theta = 52.4°$.

As θ is varied close to the direction for SCPM, simultaneous phase matching of the same two processes also occurs in a noncollinear fashion with radiation at the common wavelength having a wave vector $\vec{k}_1^o$ shared by both processes. This situation is shown in Fig. 1(b) where families of curves for various values of θ for the two processes are given. The noncollinear phase-matching angles ψ and β (measured internal to the crystal) are plotted as a function of λ_1^o. ψ is the angle between the pump direction and $\vec{k}_1^o$, and β is the angle between the pump direction and the upconverted signal. The dotted line indicates the locus of points which satisfy the simultaneous (noncollinear) phase-match conditions. The phase-matching curves for $\theta = 52.4°$ are not shown; however, they would just intersect at $\psi = \beta = 0$, i.e., where the dotted curve meets the abscissa. Thus, for $\theta < 52.4°$ the two processes cannot be phase matched simultaneously for any value of ψ, as indicated, for example, by the nonintersecting curves for $\theta = 52°$. For $\theta > 52.4°$ simultaneous phase matching is possible for some appropriate value of ψ as is evident from the figure. The curves in Fig. 1(b) are for ψ variations in the plane perpendicular to the plane defined by the c axis of the ADP crystal and the pump direction. Similar curves are obtained for ψ variations in other nonperpendicular planes. Hence, when $\theta = 52.4°$, upconverted radiation should be observed around 0.330 μm with any pump radiation which is not of purely ordinary or extraordinary polarization. As θ is increased by a few degrees, the upconverted radiation would remain as a result of noncollinear interaction ($\psi \neq 0$), but it should completely disappear below 52.4°. This tuning characteristic is distinguished from the single-step third-order process

$$\omega_p^o + \omega_p^e \to \omega_2^e + \omega_3^e, \tag{3}$$

which is also phase matched when processes (1) and (2) are simultaneously matched. The tuning curve for (3) is indicated in Fig. 1(a) for a 0.530-μm pump in ADP. For this process a sharp cutoff for the upconverted radiation for $\theta < 52.4°$ does not occur, but a relatively smooth variation with θ results from the adjustability of the frequencies ω_2 and ω_3 to preserve phase matching.

The pump source for the experimental investigation was a doubled neodymium glass laser. At the entrance face of the ADP crystal the pump had an intensity of approximately 1 MW in an area of 1×2 mm^2. The detector system consisted of several glass filters, a monochromator, and an Amperex 56UVP photomultiplier. It had a spectral bandwidth of 32 Å and subtended a solid angle of 4.7×10^{-3} sr; the observed radiation at ω_3 had approximately the same bandwidth. The pump was polarized at 45° to the plane of the c axis and the pump direction, and had a full-

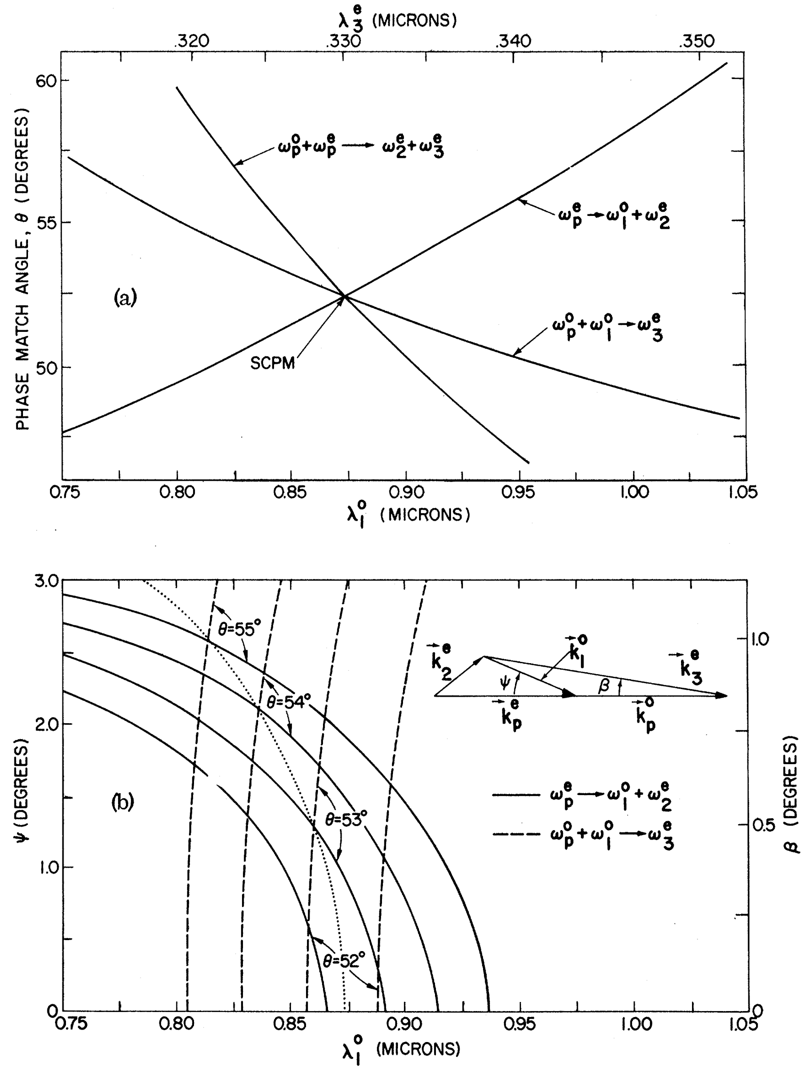

FIG. 1. Parametric processes in ADP employing a doubled neodymium laser pump for the indicated e and o polarizations of the propagating waves. (a) Tuning curves of processes (1), (2), and (3) in the text, each collinearly phase matched. The internal phase-matching angle θ is plotted as a function of the common wavelength λ_1^o (corresponding to the frequency ω_1^o) and λ_3^e (corresponding to ω_3^e). (b) The internal angles ψ and β are plotted as a function of λ_1^o for noncollinear phase matching of processes (1) and (2) for various angles θ. The dotted line gives the locus of points for simultaneous noncollinear phase matching of (1) and (2).

angle beam divergence of approximately 5 mrad. The ADP crystal was 5 cm in length. It was cut with faces normal to the direction given by θ approximately equal to 52° and an azimuthal angle of 22.5°. This orientation optimizes the SCPM process.[5] For this arrangement the effective length of the ADP is determined by walkoff and is approximately 1 cm.

Various checks of the polarization characteristics indicated in (1) and (2) were initially performed. First, the common signal ω_1^o was detected only for an extraordinary pump and it was found to have the expected ordinary polarization. Second, the upconverted signal was of extraordinary polarization and was present only for a pump with both polarization components. These polarization dependences lent additional support to the measurements which followed. In addition, the observed radiation at ω_3^e satisfied a square-law dependence on power with the pump, as anticipated from the linear dependence of the intensity of ω_1^o on pump intensity.

The results of the measurement of the θ dependence of the upconverted radiation at 0.330 μm are indicated in Fig. 2. Since the crystal was oriented to approximately 1°, the abscissa

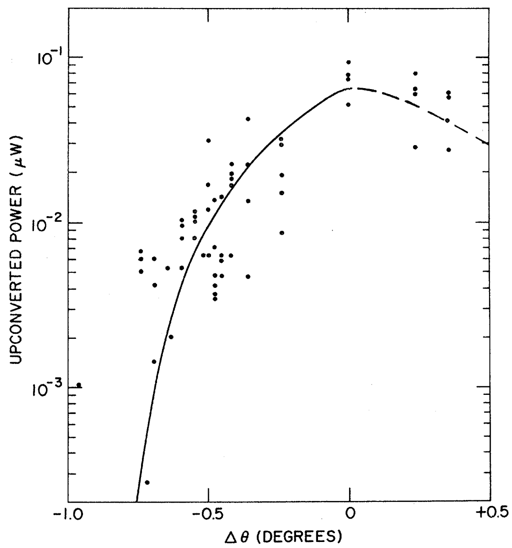

FIG. 2. Upconverted power (within the detector bandwidth of 32 Å) at 0.330 μm as a function of angular difference of θ for a doubled neodymium pump in ADP. $\Delta\theta = 0$, corresponding to $\theta = (52.4 \pm 1.0)°$, is located at the orientation for maximum signal output. The data are normalized for fluctuations in peak pump power on successive shots.

gives a relative measure $\Delta\theta = \theta - (52.4 \pm 1.0)$, where $\Delta\theta = 0$ is assigned to the orientation of maximum output. The curve roughly indicates a possible fit to the experimental points which show a large scatter. This scatter is attributed primarily to variations of the pump mode structure on successive laser shots and mechanical vibration of the crystal with respect to the pump axis. The exact shape of the curve for $\Delta\theta > 0$ depends on many experimental factors such as pump beam area, pump divergence and bandwidth, and detector bandwidth. This portion of the curve in Fig. 2 is indicated by a dashed line and in general it decreases rather slowly as compared with the $\Delta\theta < 0$ side of the curve, the latter region being dominated by the inability to phase match processes (1) and (2) simultaneously as was anticipated above. The exact theoretical curve is a complicated function of many parameters and no attempt has been made to calculate its shape. It is possible, however, to compare the magnitude of the maximum observed upconverted signal, approximately 0.1 μW, with that anticipated from theory.[1,2] The latter yields a value of approximately 0.04 μW for the experimental conditions employed in the present experiment. In addition the measured decrease of 2.5 orders of magnitude in upconverted power for a variation in the angle θ of about 0.7° is at least as great as the estimated decrease over this angle due to the falloff of the downconversion process. Thus, the data are in reasonable quantitative agreement with theoretical estimates.

The results of the present experiment indicate further that the upconverted signal could be appreciably enhanced by enclosing the nonlinear crystal in an optical cavity. The behavior would be similar to the usual optical parametric oscillator but here the phase-matched processes (1) and (2) would yield an upconverted frequency. This configuration also provides for some tunability.

In conclusion, a two-step nonlinear optical interaction has been observed. The particular processes involved provide for a direct measure of the spontaneous emission (noise) associated with the upconversion process. The magnitude of the upconverted signal is in reasonable agreement with the estimated value. This agreement tends to confirm the earlier conclusions [1,2] that in most cases upconversion noise due to this two-step process can be made negligibly small as a result of the large momentum mismatch in the spontaneous downconversion process. It is clear from the present work, however, that due care must be exercised to avoid the special cases where this need not be the situation. Finally, simultaneous collinear phase matching has been shown to be possible in several crystals at frequencies for which intense coherent sources are available. Simultaneous phase matching of different nonlinear optical processes with the coupling of one or more common waves between them would appear to open up additional possibilities for combined optical effects not previously observed.

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[1]H. A. Smith and C. H. Townes, in *Polarisation, Matière et Rayonnement, Volume Jubilaire en l'Honneur d'Alfred Kastler*, edited by the Société Française de Physique (Presses Universitaires de France, Paris, 1969), p. 467.

[2]C. L. Tang, Phys. Rev. 182, 367 (1969).

[3]J. E. Midwinter and J. Warner, J. Appl. Phys. 38, 519 (1967).

[4]ADP refractive index data were taken from F. Zernike, Jr., J. Opt. Soc. Amer. 54, 1215 (1964).

[5]J. E. Midwinter and J. Warner, Brit. J. Appl. Phys. 16, 1135 (1965).