BACKWARD-WAVE MEDIUM INFRARED DOWN-CONVERSION IN PROUSTITE

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Phase-matched backward-wave down-conversion has been obtained in the *medium infrared* using proustite (Ag_3AsS_3) as the mixing crystal. A transmission window between the two-photon lattice band and the high-frequency wing of the reststrahlen permitted 18.6 μm infrared to be generated by mixing the output of a ruby laser with stimulated Raman emission from bromoform ($CHBr_3$).

From the earliest days of non-linear optics it has been appreciated [1] that for down-conversion,

$$\nu_p - \nu_s = \nu_i,$$

phase-matching is in principle possible with the generate wave (ν_i) propagating in the opposite direction to the two driving waves (ν_p and ν_s);

$$k_p - k_s = -k_i.$$

The requirement on the crystal for this *backward-wave* phase-matching is approximately given by

$$\frac{\lambda_i}{\lambda_p}\frac{B}{\eta} \geqslant 2,$$

where B is the birefringence and η average refractive index. Because of this stringent condition, the only previously published experiments on backward-wave down-conversion have involved λ_i in the *millimetre* [2] or *far-infrared* [3] region. We report here backward-wave down-conversion to the *medium infrared*, the mixing crystal used being proustite [4, 5]. A significant transmission window (see fig. 1) between the region of two-phonon lattice absorption and the high-frequency wing of the reststrahlen [6] permits a sufficiently large ratio λ_i/λ_p to be obtained for backward-wave phase-matching.

The refractive indices of proustite have been measured by Hobden [7] for wavelengths spanning the whole range of interest, his results being presented in the form of "Sellmeier" equations for the ordinary and extraordinary indices. Using these equations it can be shown that backward-wave phase-matching cannot quite be satisfied within the normal transmission range, limited by the band-edge at short wavelengths and by a two-phonon band at long wavelengths. However, a region of limited transmission between the two-phonon-band and the reststrahlen does permit backward-wave phase-matching. The effective non-linear susceptibility is largest for type I matching,

$$k_p^e - k_s^o = -k_i^o;$$

that is to say with the infrared generated generated as an ordinary wave. The absorption of proustite is plotted in fig. 1 for the ordinary polarisation and wavelengths within the two-phonon/reststrahlen window. In the experiment reported below λ_i was located at the 18.6 μm absorption minimum, the coefficient α_i^o then being 10.5 cm^{-1}. The phase-matching angle calculated from Hobden's equations is 65.4° for λ_i = 18.6 μm, λ_p = 694 nm and with the small non-collinearity discussed below taken into account. The useful window, defined by $\alpha^o \leqslant 20$ cm^{-1}, extends from 15.2 to 20.8 μm; the forward phase-matching condition is satisfied throughout the window but backward-wave matching is only possible for wavelengths longer than 15.9 μm. The validity of Hobden's "Sellmeier" equations for the two-phonon absorption region have previously been con-

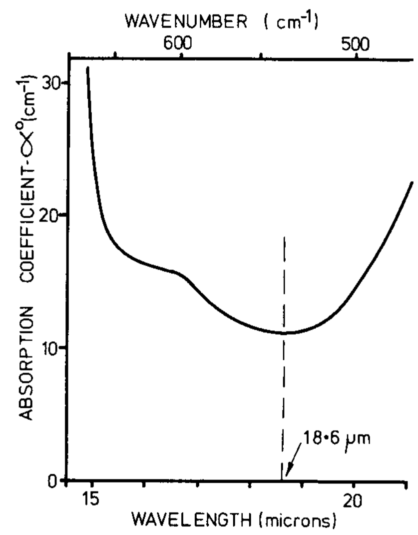

Fig. 1. Infrared absorption for proustite with ordinary wave polarisation.

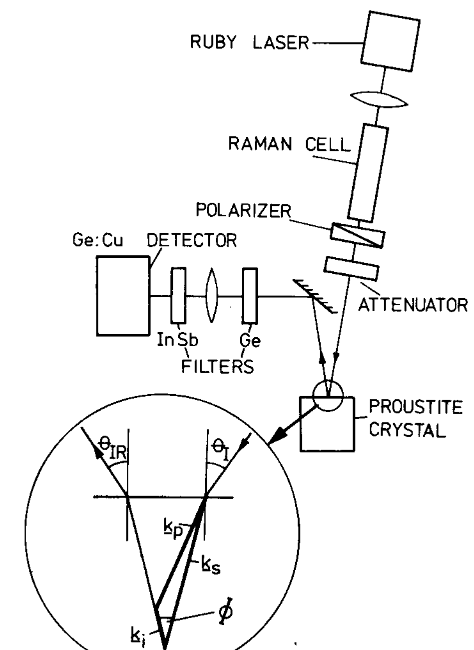

Fig. 2. Schematic arrangement for observing backward-wave down-conversion.

firmed by experiments [8] on parametric fluorescence in proustite generated by a ruby laser.

For our experiment the high frequency waves were provided by a ruby laser (694 nm) and stimulated Raman emission from bromoform [9] ($\bar{\nu}_v$ = 539 cm^{-1}, λ_{Stokes} = 721 nm). Although the experimental arrangement (fig. 2) ensured that the two beams were collinear before entering the crystal, the large birefringence of proustite resulted in significantly different angles of refraction for the ordinary and extraordinary components and hence some non-collinearity within the crystal. The backward-wave phase-matching was therefore slightly non-collinear (see fig. 2) with the angle $\Phi \simeq 10°$. An interesting feature of backward-wave non-collinear phase-matching is that when angles are small enough for $\sin x \simeq x$, the generated infrared leaves the crystal at an angle equal to the reflection angle for the input beams ($\theta_{IR} = \theta_I$).

There are two important considerations concerning the acceptable mis-match $|\Delta k|$. In the first place, eq. (1) of Yang et al. [3] shows that for crystals with absorption lengths (α_i^{-1}) much smaller than the physical length of the crystal

$$|\Delta k(\text{fwhm})|\alpha_i^{-1} = 1 .$$

Secondly the expression relating the down-conversion band-width $\Delta\bar{\nu}$(fwhm) to $|\Delta k|$ contains in the denominator sums of indices and dispersions, and not differences as for the forward-wave case, the result being a much smaller band-width for a given set of frequencies. Thus

$$\Delta\bar{\nu} = \frac{|\Delta k|}{2\pi[\eta_s + \eta_i + \nu_s(\mathrm{d}\eta_s/\mathrm{d}\nu_s) + \nu_i(\mathrm{d}\eta_i/\mathrm{d}\nu_i)]}$$

and inserting $|\Delta k| = \alpha_i^o = 10.5$ cm^{-1}, together with appropriate indices etc., gives a $\Delta\bar{\nu}$ equal to 0.27 cm^{-1}.

A simple arrangement was used for the experiment. The dye *Q*-switched ruby laser yielded ~ 1.5 MW in an essentially TEM_{00} mode and with a spectral spread < 0.1 cm^{-1}. The ruby laser beam was focused by a 0.3 m lens through a 0.5 m cell of bromoform, a first Stokes power of ~ 300 kW being produced with pulse shortening from 7 to 5 ns. The stimulated Raman band-width was narrowed to ~ 2 cm^{-1} from the 4 cm^{-1} spontaneous width (both fwhm). Both the ruby and Raman beams were polarised in a plane perpendicular to that containing the propagation direction and the crystal optic axis. So as to provide the ordinary and extraordinary components required for phase-matching, a polariser was placed after the bromoform cell set at 45° to the initial polarisation direction. An attenuator was also required to keep power densities below the damage threshold of proustite. The combined transmission of the polariser plus the attenuator was 20%, and the beams had a diameter of 2 mm at the crystal. The crystal used* had been cut for type I phase-matching in a quadrant for which the non-linear susceptibility elements $|d_{31}|$ and $|d_{22}|$ are additive. A Ge:Cu infrared detector at 4.2 K was used with visible/near-infrared rejection provided by Ge and InSb filters. A grid polariser was employed for checking the polarisation of the 18.6 μm radiation.

The infrared power generated by backward-wave down-coversion was ~ 5 mW. It was confirmed that the polarisation selection conditions for type I matching were obeyed and that the internal phase-matching angle was within 0.25° of that calculated above. From the discussion earlier, it is apparent that the Raman band-width was at least five times larger than the bandwidth for down-conversion. Calculations taking this effect into account yield a predicted infrared power about an order of magnitude larger than that measured. In addition the measured external angular range of ~ 1.5° for phase-matching was significantly greater than the predicted value of ~ 0.7°.

To summarise, we have demonstrated backward-wave down-conversion to the *medium infrared*. The idea of using the transmission window between the two-phonon band and the reststrahlen offers a tuning range for proustite of ~ 175 cm^{-1} centred on ~ 550 cm^{-1}. It is envisaged that similar windows can be exploited in other materials, possibly for even longer wavelengths.

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