TEMPERATURE-DEPENDENT PHASE MATCHING FOR FAR-INFRARED DIFFERENCE-FREQUENCY GENERATION IN InSb

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The efficiency of difference-frequency generation in InSb at 100 cm^{-1}, using a CO_2 laser as input, was found to be strongly temperature dependent. This dependence is shown to be due to phase matching resulting from an increase with temperature of the short-wavelength refractive indices. A value of 4.03 is derived for the refractive index at 100 μ. The effect of astigmatism in the input optics is pointed out.

In an earlier experiment[1] a far-infrared frequency (101 cm^{-1}) was generated as the difference between two frequencies (946 and 1047 cm^{-1}) emitted by a single Q-switched CO_2 laser.[2] The nonlinear material was a 1-mm-thick slab of InSb cut in the (110) plane and cooled to 77°K. This Letter describes subsequent experiments in which it was observed that the signal from thicker crystals vanishes at 77°K but has a strong maximum at higher temperatures.

The experimental setup is as follows: The laser has a nearly hemispherical cavity 138 cm long and is Q switched at 1600 pulses/sec. The gas mixture consists of approximately 22 Torr He, 2.8 Torr CO_2, and 2.8 Torr N_2. The discharge is 1 m long and the current is 30 mA. The laser oscillates at a number of lines; however, sum-frequency measurements show that only one line, at 1047 cm^{-1} ($P20$), seems to mix with either one at 946 cm^{-1} ($P18$) or one at 944 cm^{-1} ($P20$). The beam from the laser is focused on the mixing crystal by a mirror with a focal length of 110 cm. The polarization of the beam is parallel to the [111] direction in the crystal. A spherical mirror refocuses the output into a light pipe leading to the liquid-helium-cooled Ge:Ga photoconductive detector.[3] A crystal-quartz window at the opening of the lightpipe absorbs the radiation from the laser. The signal from the detector is processed in the normal way using a boxcar integrator synchronized to the Q-switched pulse. The InSb used had the following properties: p-type, $\mu = (7.3\text{-}7.6)\times 10^3$ cm^2 V sec^{-1}, and $n_e = (1.8\text{-}2.6)\times 10^{14}$ cm^3 (77°K).

Figure 1 shows curves of signal versus temperature for three different crystal thicknesses. The appearance in a mixing experiment of a temperature-dependent maximum, such as shown in these curves, immediately leads to the supposition that this behavior is caused by a change with temperature of the refractive indices, giving perfect phase matching at the temperature at which the maximum occurs. However, if only the real part of the indices would change with temperature, the maximum would occur at the same temperature for all thicknesses and the signal at the maximum would be proportional to the square of the crystal thickness. Figure 1 shows that this is not so: The maxima do not occur at the same temperature and the signals at the maxima are not proportional to the square of the crystal thicknesses. Clearly both of these features are due to the fact that the absorption increases rapidly with temperature, especially at 100 μ, as is shown in the dashed curve in Fig. 1.

To substantiate this temperature-dependent phase-matching argument, the refractive indices at 946 and at 1047 cm^{-1} were measured as a function of temperature; they are shown in Table I. Also shown in Table I is a value $n_3' = n_2 + (n_1 - n_2)\nu_1/\nu_3$ which is the index value required for perfect phase matching at the difference frequency ν_3.

To arrive at a value for the refractive index at ν_3 it was assumed that in the absence of absorption, perfect phase matching would occur at a

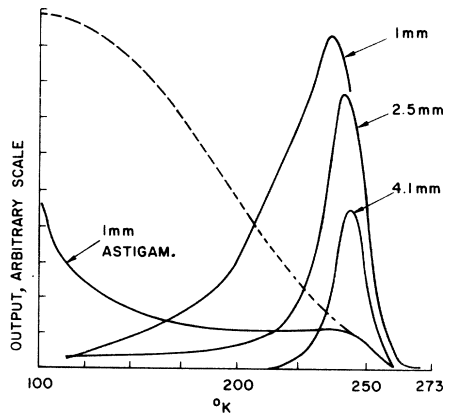

FIG. 1. Signal versus temperature for three different crystal thicknesses. The dashed curve shows transmission versus temperature at 100 μ.

Table I. Refractive indices as a function of temperature.

T (°K)	n_1 (1047 cm^{-1})	n_2 (946 cm^{-1})	n_3'
75	3.854	3.848	3.903
100	3.862	3.855	3.918
125	3.873	3.868	3.935
150	3.886	3.880	3.954
175	3.890	3.892	3.976
200	3.916	3.906	3.998
225	3.930	3.921	4.020
250	3.944	3.933	4.044
275	3.956	3.943	4.074
300	3.965	3.952	4.093

temperature slightly higher than the temperature at which the maximum for the 4.1-mm-thick crystal occurs, i.e., at 245°K, giving $n_3 = 4.039$.

The change with temperature of the short-wavelength indices is large because of the nearness of the absorption edge and its temperature shift. However ν_3 is far removed from the nearest Reststrahlen band (183 cm^{-1}) and moreover the position of this band changes only little with temperature.[4] Thus it may be assumed, reasonably, that n_3 does not change with temperature, and on this assumption the momentum mismatch at temperatures other than 245°K may be calculated. Using these data in Eq. (1) the temperatures at which the signal becomes vanishingly small for the different thicknesses may be determined. This gives 217°K for 4.1 mm, 198°K for 2.5 mm, and 125°K for 1 mm. These values agree fairly well with Fig. 1.

The refractive index at 100 μ was also measured by mixing in a prism (angle 12°48′). Both inputs were incident at 90° to preserve their collinearity inside the crystal. The exit angle for ν_1 at 243°K was 63°15′ ± 15′, giving $n_1 = 4.03 \pm 0.01$.

Assuming single-mode plane waves of uniform intensity at the input frequencies, the angular dependence of the output can be shown to be

$$D = \left[\frac{2J_1(k_3 d \sin\varphi)}{k_3 d \sin\varphi}\right]^2 L \qquad (1)$$

with

$$L = e^{-\alpha_3 l}\,\frac{1+e^{-\Delta\alpha l}-2e^{-\frac{1}{2}\Delta\alpha l}\cos[k_3(1-\cos\varphi)+\Delta k]l}{(\frac{1}{2}\Delta\alpha)^2+[k_3(1-\cos\varphi)+\Delta k]^2}.$$

Here $\Delta\vec{k} = \vec{k}_1 - \vec{k}_2 - \vec{k}_3$, where $\vec{k}_{1,2,3}$ are the respective propagation vectors at the three frequencies, d is the radius of the interaction cylinder, φ is the angle between the axis of the input beam and the direction of the output radiation, l is the crystal thickness, and $\Delta\alpha = \alpha_1 + \alpha_2 - \alpha_3$, where $\alpha_{1,2,3}$ are the absorption coefficients at the three frequencies. For $\Delta k = 0$ and no absorption[5] L reduces to the familiar $(\mathrm{sinc}\, y)^2$ function with $y = \frac{1}{2}k_3 l(1-\cos\varphi)$.

For the values of l (1 mm; $2\frac{1}{2}$ mm) and d (1 mm) used here, the value $\varphi = \varphi_0$ for which the output is zero is determined by the first term on the right in Eq. (1). Since this term is the diffraction pattern of a circular aperture of radius d at the difference frequency ν_3 and does not contain l, φ_0 should be the same for both thicknesses. From Eq. (1) one calculates $\varphi_0 = 3.5°$. This is confirmed by the experimentally measured output distributions shown in Fig. 2.

As shown in Fig. 1, no signal was obtained from the 1-mm-thick crystal at temperatures below 100°K. This would seem to be inconsistent with the earlier experiments mentioned above, where a signal larger than the maximum signal shown in Fig. 1 was obtained at 77°K.[1] The only difference in the previous experimental setup was that the inputs were focused by a mirror with 27-cm focal length which was used rather far off axis. This introduces astigmatism which apparently increases the coherence length in the

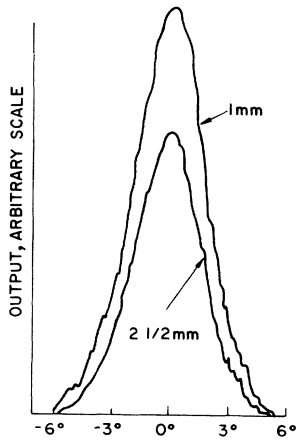

FIG. 2. Angular distribution of the output. Instrument width 2°.

1-mm crystal at 77°K. A curve of signal versus temperature obtained with a 1-mm-thick crystal in this configuration is shown in Fig. 1. Note that this curve shows a much smaller signal at the phase-matching temperature than the corresponding curve with the large-radius mirror. The angular dependence of the output and the variation of the signal with temperature were shown to be very dependent upon the position of the crystal with respect to the horizontal and vertical focal lines of the off-axis mirror. A detailed treatment of this subject will be given separately.

Since the absorption at all three frequencies increases rapidly with temperature it is obviously preferable to phase match at a low temperature. A method of doing this in the collinear case using magnetoplasma effects has recently been described by Van Tran and Patel.[6] Since the index in the far infrared is larger than that in the near infrared, phase matching can also be obtained by having the input beams incident at a small angle to each other. In a first experiment utilizing this method a power in excess of 1 μW was obtained at 106 cm^{-1} by mixing two separate Q-switched CO_2 lasers incident at an angle, inside the crystal, of approximately 2° to each other. A full account of these experiments will be given at a later date.

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STRESS-INDUCED EXCHANGE SPLITTING OF HYPERBOLIC EXCITONS IN GaAs.*

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Evidence for the existence of hyperbolic excitons has been obtained from a study of the effects of compressive uniaxial stresss on optical structure of GaAs at 77°K using a double-beam wavelength-modulation technique. We have observed a polarization-dependent splitting of this structure which cannot be accounted for on the basis of one-electron band theory but is explained by including the electron-hole exchange interaction. An estimate of an effective radius of ≈10 Å for the electron-hole interaction has been made.

The existence of significant contributions of the electron-hole Coulomb interaction (i.e., exciton effects) to the experimental optical spectra in the vicinity of M_1 (hyperbolic) interband transitions [for example, the E_1-$(E_1+\Delta_1)$ structure in diamond- and zinc-blende-type semiconductors] has been suggested by several authors.[1-6] The experimental evidence is not conclusive although a recent study of the wavelength derivative spectra of the E_1-$(E_1+\Delta_1)$ structure in InSb strongly suggests that the electron-hole Coulomb interaction must be taken into account.[7] In this paper we present conclusive evidence for the existence of hyperbolic excitons based on symmetry considerations and not dependent on interpretations of line shape.[7] The E_1-$(E_1+\Delta_1)$ structure of GaAs has been studied as a function of static uniaxial compression along [100] at 77°K using a double-beam wavelength-modulation technique. One-electron band theory predicts that no splitting for this optical structure should occur for [100] stress since for this stress direction the valley degeneracy is not changed[8] and stress does not remove the Kramers' degeneracy.[9] However, a small polarization-dependent splitting is observed and is explained by including the electron-hole exchange interaction as well as the one-electron deformation potential. From these measurements the effective radius for the electron-hole interaction is estimated to be ≈10 Å.

Several optical-modulation methods (e.g., electroreflectance,[10,11] piezoreflectance[12]) have been used to study critical-point structure in the optical spectra of semiconductors. Of these methods, wavelength-modulated reflectance (WMR) is particularly attractive from a theoretical