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Nonlinear Optical Properties of $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ in the Tetragonal Phase*

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Geusic *et al.*¹ have reported the existence of large nonlinear optical coefficients combined with the absence of optical damage in the material $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$. Smith *et al.*² have used this material to attain low threshold cw optical parametric oscillation. In this letter, we report on the properties of $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ between 300° and 560°C, where it exists in a ferroelectric tetragonal phase. In this region, the crystal is uniaxial, does not require detwinning, and is of higher optical quality than below 300°C. Also, since the half-wave voltage is considerably lower than at room temperature it should be possible to achieve significant electro-optic tuning of optical parametric oscillators. We note that some measurements in the tetragonal phase have been reported by Singh *et al.*³

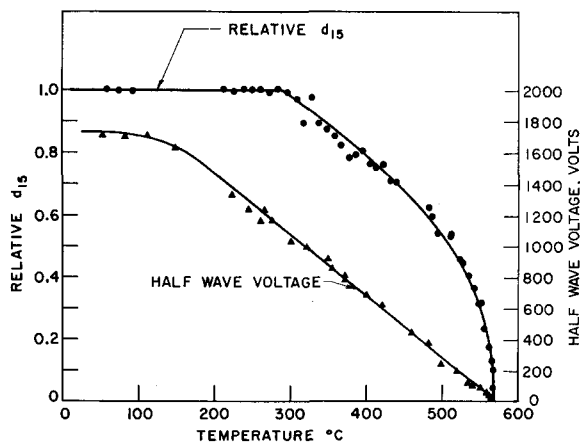


FIG. 1. Optical nonlinearity and half-wave voltage as a function of crystal temperature.

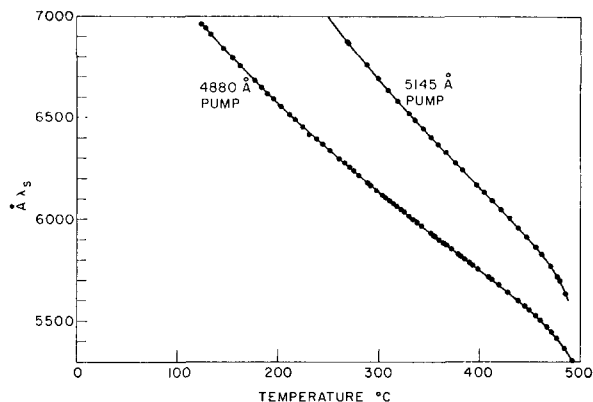


FIG. 2. Wavelength of spontaneous parametric emission as a function of crystal temperature.

The variation with temperature of the nonlinear coefficient $d_{15}=d_{31}$ was determined by SHG with a continuously pumped 1.06μ Nd:YAG laser. The second-harmonic power was recorded while continuously scanning the crystal temperature from room temperature to 600°C, producing the characteristic $(\sin x/x)^2$ curve. As the temperature was raised from room temperature, the peak height of the side lobes built up and reached a maximum at the phase-matching temperature of $T=116^\circ\text{C}$. About 25 peaks of the second-harmonic intensity were observed between 40° and 116°C, and 320 peaks with successively declining amplitudes were observed from there to 580°C. The relative magnitude of d^2 was calculated by noting that the ratio of the second-harmonic power generated at the peak of the n th side lobe to that generated at the phase-matching temperature is given by

$$(d^2/d_0^2)/[(n+\frac{1}{2})\pi]^2,$$

where d is the magnitude of the optical nonlinearity at the temperature of the n th side lobe, d_0 is its magnitude at the phase-matching temperature, and $n \geq 1$. As shown in Fig. 1, the nonlinearity is constant from room temperature to about 300°C, and then breaks sharply and decreases to zero at the Curie temperature of $\sim 560^\circ\text{C}$. It should be noted that the $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ crystals⁴ were striated and remained so over the full temperature range examined. However, above 300°C a significant reduction of crystal strain was observed.

The half-wave voltage at 6328 Å as a function of temperature is also shown in Fig. 1. The data were taken by applying an electric field along the c axis and measuring the voltage necessary to cause a shift of one fringe for light propagating down the a axis.

The potential tuning curves for an argon-pumped- $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ parametric oscillator are shown in Fig. 2. The data was taken by observing the wavelength of on-axis spontaneous parametric emission as a function of crystal temperature.⁵ The tuning curves are smooth and continuous through the phase transition at 300°C. The spontaneously emitted power decreases rapidly just below 500°C as the idler enters a region of high loss and rapidly increasing dispersion. Extrapolation from these curves indicates that the visible portion of the tuning curve for a doubled YAG pump would lie in the tetragonal-phase temperature range for 90° phase matching.

Calculations indicate that it should be possible to electro-optically tune⁶ an optical parametric oscillator about 1000 cm^{-1} by using $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ at a temperature of 550°C, where the half-wave voltage is about 100 V and the optical nonlinearity is still reasonably large. However, as a result of the low resistivity (about $1.3 \text{ M}\Omega\cdot\text{cm}$) at these temperatures, such tuning could

probably be achieved only on a repetitively pulsed basis if excessive crystal heating is to be avoided.

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Photoemission from *p*-GaSb Treated with Cesium and Oxygen

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The photoemission from cesiated GaP¹ and GaAs² and cesium-oxygen-treated InP,³ all demonstrate emission of electrons from *p*-type III-V compounds when the surface work function is equal to or less than the bandgap energy. In this case the photoelectric threshold is determined by the bandgap energy. In order to observe the effect of the addition of cesium or cesium plus oxygen for the case where the photoelectric threshold is determined by the reduced surface work function, it was necessary to study a material with a bandgap energy somewhat below the expected reduced work function. The result for such a material, *p*-GaSb, with a bandgap of about 0.7 eV is reported.

The experiments were conducted with highly doped *p*-type GaSb ($\mu_H = 150 \text{ cm}^2/\text{V}\cdot\text{sec}$, $\rho = 7.8 \times 10^{-4} \Omega\cdot\text{cm}$, $N = 5.3 \times 10^{19} \text{ Zn}$). The crystal was cleaved on the [110] face in ultrahigh vacuum ($< 10^{-10}$ Torr).

The experimental apparatus is shown schematically in Fig. 1. Cesiumation was accomplished with a cesium-ion source so that the cesium arrival rate to the crystal could be monitored. The cesium-ion gun was formed from nickel tubing filled with a mixture of cesium chromate and silicon, which when resistively heated

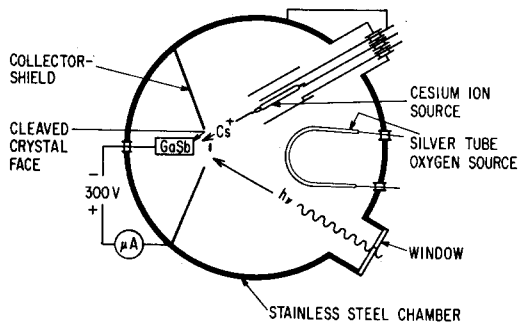


FIG. 1. High-vacuum chamber with provisions for cleaving, cesiating, and oxygen treatment.

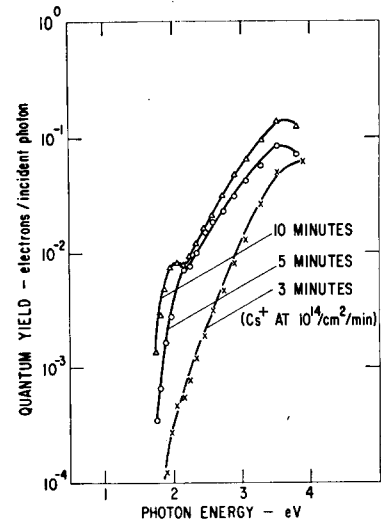


FIG. 2. Electron yield vs photon energy showing the progressive effects of cesiation.

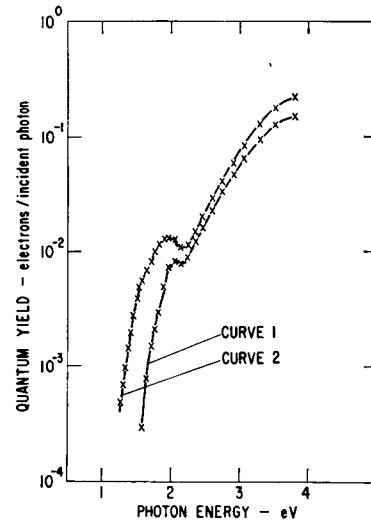


FIG. 3. Electron yield vs photon energy showing the effect of oxidation.

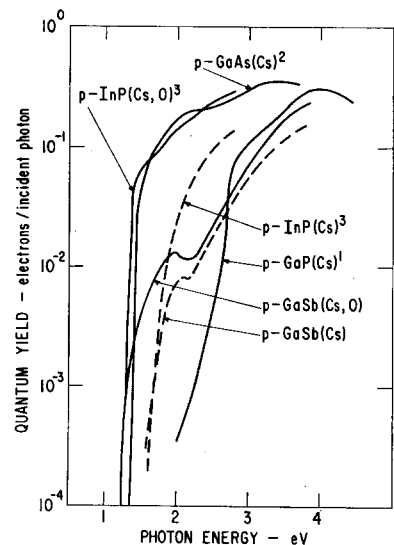


FIG. 4. Electron yield of various *p*-type III-V compounds.