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**Demonstration of Temperature-Insensitive Harmonic Generation
in Deuterated L-Arginine Phosphate**

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The thermo-optic coefficients of deuterated l-arginine phosphate indicate temperature-insensitive phase-matching directions for second-harmonic generation. We present direct measurements that partially agree with these predictions, and provide an explanation of the discrepancies between predicted and measured behavior.

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Efficient frequency conversion of high power laser radiation requires materials that possess significant nonlinear coefficients, high damage threshold, and the capability to phase-match, with reasonable angular bandwidth, the interaction between the fundamental and harmonic waves. Deuterated L-arginine phosphate (d-LAP) is a very promising new crystal for use in harmonic frequency conversion devices for Nd³⁺ lasers.¹ In addition to having a larger nonlinearity than KDP, d-LAP possesses another property that is quite attractive for high average power applications: the existence of temperature-insensitive phase-matching directions. In general, biaxial materials, such as d-LAP, are far more likely to demonstrate temperature-insensitive phase-matching directions than uniaxial crystals, such as KDP. Biaxial materials must satisfy far less stringent relationships between refractive indexes and thermo-optic coefficients than uniaxial materials in order to exhibit this property. For efficient high average power frequency conversion, careful management of the thermal loading of the conversion crystal is essential.² The presence of phase-matching directions with large thermal bandwidth greatly eases the requirements for crystal temperature control.³

In the principle planes, the reported thermo-optic coefficients of d-LAP¹ predict relatively large changes in the phasematching angle as a function of temperature for both type I and II phase-matching processes. Furthermore these rates of phase-matching angle change have opposite signs in the two principle planes. This behavior indicates the existence of temperature-insensitive phasematching directions.

Utilizing a modelocked, Q-switched Nd³⁺:YAG laser and a very high quality, spherically shaped sample of d-LAP mounted in a goniometer, we measured directly the change in phase-matching angle as a function of temperature in the principle planes. The values we have obtained, shown in Table 1, from this procedure directly indicate the presence of temperature-insensitive directions for type I phase-matching of second harmonic generation of 1.064 μ m light. Using this same apparatus, we have also accurately mapped the entire phase-matching locus for both type I and II second harmonic generation. By fitting these locus maps to a very simple analytical approximation, we have been able to predict the approximate location of the temperature-insensitve directions for type I phase-matched second harmonic generation.

Although our measurements do indicate temperature-insensitive phase-matching directions in the type I second harmonic generation locus, the values we measure for the rate of change of phase-matching angle with respect to temperature in the principle planes disagree significantly with the predictions made from the early thermo-optic data¹. Furthermore, our direct measurements indicate that there are no temperature-insensitive phase-matching directions for type II second harmonic generation. The source of this discrepancy lies in the highly anisotropic thermal expansion of d-LAP,

which requires the thermo-optic coefficients obtained from minimum deviation measurements be corrected for thermally-induced changes in the prism angles of the samples.

References:

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Table 1. Measured values of the rate of change of phase-matching angle with respect to temperature in the principle planes.

nonlinear process	measured value of $d\theta_{PM}/dT$ ($\mu\text{rad}/^\circ\text{C}$)	
	α - β plane	α - γ plane
Type I SHG	$+248 \pm 4$	-38.3 ± 1.9
Type II SHG	$+237 \pm 4$	$+30.9 \pm 2.0$