This Week's Citation Classic

The coefficients that describe optical second harmonic generation in piezoelectric crystals are shown to be equal to a triple product of linear susceptibilities times an element of a third rank tensor which has the same value for all crystals so far investigated. [The *SCI*® indicates that this paper has been cited over 225 times since 1964.]

Robert C. Miller
Bell Laboratories
Murray Hill, NJ 07974

October 6, 1980

"The pioneering demonstration by PA. Franken et al. in 1961 of optical second harmonic generation in quartz using a ruby laser[1] started modern nonlinear optics. The phenomena of interest at that time involved laser induced optical polarization fields that were quadratic in the incident laser electric field and therefore required piezoelectric crystals, i.e., crystals that lack a center of inversion. With a background of research in ferroelectric crystals, a subclass of piezoelectric crystals, which resulted in a rather large collection of ferroelectric crystals and the ready availability of lasers at Bell Laboratories, I concluded that there were a number of interesting nonlinear optical experiments that I could perform with a modest investment in time and equipment.

"The strength of the nonlinear interaction of the optical beams with the nonlinear optical materials is determined by an element of a third rank tensor, d, but which crystals will have large d's? In spite of the beautiful formal theoretical treatment of the problem by N. Bloembergen and his group,[2] the search for efficient nonlinear optical materials was by and large a hit and miss proposition. By early 1964 the magnitudes of

d had been determined for ten or so crystals (mostly by us) and found to vary from one material to another by more than two orders of magnitude. I noted that crystals which had large linear susceptibilities (indices of refraction) had large nonlinear susceptibilities (d), an observation that led me to attempt to formulate the problem from a free energy expressed in terms of optical polarizations rather than optical electric fields as had been done earlier by others.[2] With the problem formulated in this manner it was found that d could be expressed as a triple product of linear optical susceptibilities (known quantities) multiplied by an element 6 of another third rank tensor which turned out to be nearly constant for all the crystals studied. While d's varied over a range of about 600 the δ's were within a factor of two of the average value. Thus for the first time we had a simple means of estimating d. I believe this was the main appeal of the paper. The quantity δ is sometimes referred to as 'Miller's δ' and sometimes not even referenced. This principal result stimulated both experimentalists and theoreticians, the lat-ter groups to try and understand why 6 should be nearly constant.

"A paper describing the ä-formulation was submitted to Applied Physics Letters. The referee, a distinguished physicist in the field, whose name was surprisingly given to me by the editor, was somewhat negative but he had one objection that resulted in an added sentence. The obvious utility of this work must have won over the editor for in the end it was accepted with little effort on my part.

"Like all simple rules, there are bound to be shortcomings, and there have been with the δ's. The several more recent theoretical treatments of the problem are much more satisfactory from a physics point of view.[3] However, the simplicity and reasonable success of the δ-approach continues to be useful to some investigating new materials for nonlinear optics."

1. **Franken P A, Hill A E, Peters C W & Weinreich G.** Generation of optical harmonics. *Phys. Rev. Lett.* **7**:118-19, 1961.
2. **Armstrong J A, Bloembergen N, Ducuing J & Pershan P S**. Interactions between light waves in a nonlinear dielectric. *Phys. Rev.* **127**:1918-39, 1962.
3. **Lines M E & Glass A M.** *Principles and applications of ferroelectrics and related materials*. Oxford: Clarendon Press, 1977. 477 p.