

Phase-Matching in Nonlinear Optical Compounds: A Materials Perspective

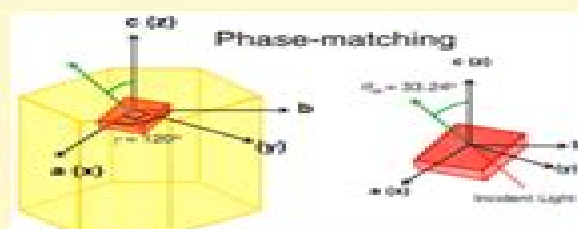
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 Supporting Information

ABSTRACT: Angle phase-matching in nonlinear optical (NLO) materials is critical for technological applications. The purpose of this manuscript is to describe the concept of phase-matching for the materials synthesis NLO community. Refractive index and birefringence are defined with respect to uniaxial and biaxial crystal systems. The phase-matching angle and wavelength range, Type I and Type II, are explained using real NLO materials, $\text{K}_2\text{B}_4\text{O}_7/\text{Cl}$ (KBOC) and $\text{Ba}_2(\text{ZnB}_2\text{O}_{10})\text{PO}_4$ (BZBP). In addition, we describe how refractive index measurements are performed on single crystals and how the resulting birefringence impacts the phase-matching. Our goal is to provide a description of phase-matching that is relevant for the materials synthesis NLO community.



INTRODUCTION

For nonlinear optical (NLO) materials, the ability to phase-match, or be phase-matchable, is of paramount importance. It is through phase-matching that NLO materials are technologically applicable. In other words, it is not enough for a given NLO material to be highly efficient at a particular wavelength; the material must also be phase-matchable. However, what exactly is the role of structure and chemistry in phase-matching? Importantly, how is phase-matching relevant to functional inorganic NLO materials?

The purpose of this manuscript is to

- (i) Describe the concept of phase-matching in powders and crystals, i.e., Type I and Type II. Refractive index, birefringence, uniaxial, and biaxial will also be defined.
- (ii) Discuss how the refractive index is measured on crystals.
- (iii) Describe how the phase-matching angle and wavelength range for uniaxial and biaxial crystals can be determined.
- (iv) Describe how the birefringence of the material impacts the phase-matching angle and wavelength range.

We will be discussing angle phase-matching (sometimes called index matching, angle matching, or birefringence matching), but hereafter simply phase-matching, that is distinct from quasi-phase-matching^{1–3} and noncritical phase-matching methods.^{4,5}

PHASE-MATCHING

The most straightforward definition of phase-matching is when the wave vectors of the fundamental radiation and second-harmonic radiation are equal. This occurs in a second-harmonic generating (SHG) material when the refractive indices of the fundamental wave and second-harmonic wave are equal, i.e., $n(\omega) = n(2\omega)$. In their seminal 1968 paper, Kurtz and Perry,⁶ and

updated 1976 paper, Dougherty and Kurtz,⁷ demonstrated that phase-matching could be determined from sieved powders. (In the 1968 and 1976 papers, the terms Type I and Type II phase-matching are not used. The phase-matching determined from powders is an average value of Type I and Type II.) If the SHG efficiency is measured as a function of particle size, one can determine if the material is phase-matchable or not (see Figure 1). It is important, however, to recognize the limitations of this measurement. A particle size vs SHG intensity measurement:

- tells you if the material is phase-matchable at a specific wavelength, the incident wavelength used.

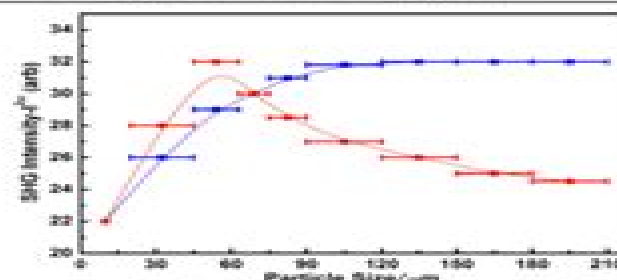


Figure 1. Phase-matching (blue) and nonphase-matching (red) curves for powder SHG measurements.

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