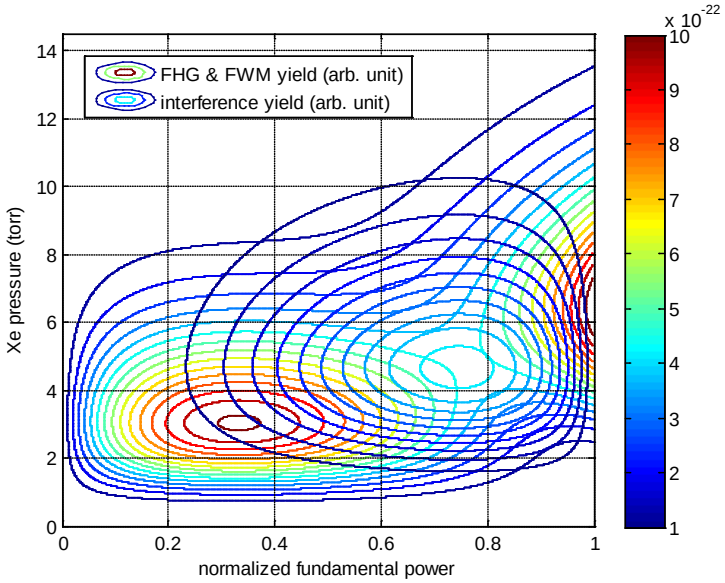
Using the formulism of perturbative nonlinear optics, we derived the general formula for generation of third- and fifth harmonic signals in inert gases by coherently controlled two-color laser pulses. There are three processes that contributed to third- and fifth harmonic signals. There are direct third- and fifth harmonic generation, four-wave mixing, and the interference between the two former processes. Key parameters affecting nonlinear conversion are phase matching or pressure of the gas, power ratio and relative phase of the two-color exciting light and focusing. The most effective way of nonlinear conversion is through optimization of the four-wave mixing process. High modulation depth of the generated harmonics is achieved by manipulating the interference term. Experimentally, the third harmonic (355 nm) signal excited by coherently controlled two-color (1064, and 532 nm) optical pulses in argon gas were found to be in good agreement with theoretical predictions.

我們理論上推導出由聚焦雙色光在各向同性介質中激發的三倍頻及五倍頻的諧波產生的通式。諧波產生主要來自三種過程的貢獻，分別為直接三或五倍頻和四波混頻激發的貢獻，以及前兩項產生過程之干涉的貢獻。我們發現在強聚焦雙色光激發的情況下，當需要高的轉換效率時，要使四波混頻產生的效率最高，而當需要較高的調制程度時，要使干涉項產生的效率最高。實驗上我成功的觀察到在氬氣中由波長 1064nm 及 532nm 激發所產生的 355 nm 訊號隨著氣體壓力以及驅動雷射強度對調制程度的影響。



P5I+ P5II /P1+P2 and P5III /P1+P2 for vary with Xe pressure and normalized fundament power at 106.4nm in the tight-focusing limit