Effects of Phase-Matching on High Harmonic Generation from Aligned N_2

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Synopsis We have used two time-delayed laser pulses to examine high harmonic generation from transiently aligned, room temperature N2 molecules in a hollow-core waveguide. The extended interaction length in the waveguide allows us to identify alignment-dependent propagation effects which mask the single molecule response. In particular, we observe a reversal in the harmonic efficiency from parallel to perpendicular alignment. This reversal depends on laser intensity and harmonic order. Simulations indicate that it can be attributed to the alignment-dependence of the tunneling ionization rate and the concomitant modification of the phase-matching conditions due to the changes in the free-electron density in the fiber.

Recently, McFarland et al [1] observed an intensity- and order-dependent reversal in the preferred alignment direction for maximal harmonic emission from N_2. This reversal was attributed to a feature of the single molecule response, HHG contributions from a more tightly bound molecular orbital. Similar reversals observed in CO_2 were also ascribed to single-molecule effects [2]We find reversals in the preferred alignment direction in N_2 which are due solely to changes in the phase-matching conditions within the target gas.

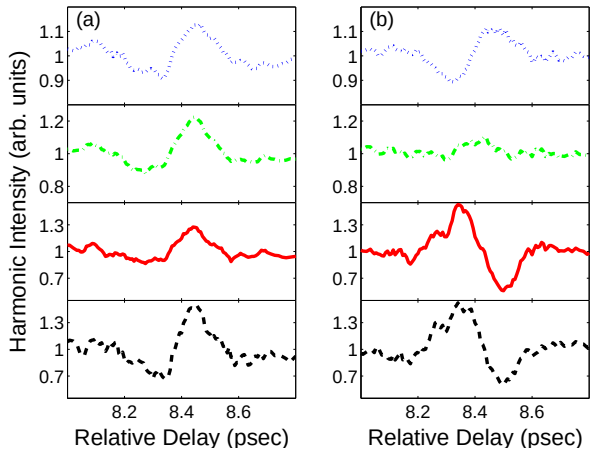

Fig. 1. Yields in the (top to bottom) 17^{th}-23^{rd} harmonics near the first alignment revival.

Figure 1 shows the yields in 17^{th} through 23^{rd} harmonic orders as a function of the delay between the signal and alignment laser pulses. These data were collected under nominally identical experimental conditions except that the signal pulse duration(intensity) was 10% smaller(greater) in Fig. 1b. The variations in the harmonic yield observed in Fig. 1a are in good agreement with predictions based on the single molecule response, assuming ionization and recombination from the HOMO. As shown in Fig. 1b, under slightly different experimental conditions, the preferred alignment direction reverses for higher orders, indicating a change in the single molecule response and/or macroscopic propagation effects.

The reversals in the harmonic yield also depend on the N_2 target gas pressure. At low laser intensity and low harmonic orders, harmonics are more efficiently generated from parallel rather than perpendicularly aligned molecules, regardless of pressure. However, at slightly higher laser intensity, the yields in the higher order harmonics are greater for perpendicular alignment over a range of pressures. The pressure dependence of the reversal provides clear evidence that the phenomenon we observe is due, at least in part, to propagation rather than single-molecule effects. Simulations that include the ionization rate anisotropy and its effect on phase-matching through the alignment-dependent free electron density are in qualitative agreement with our observations, suggesting that the reversal we observe is due solely to macroscopic propagation effects. This work is supported by DOE BES.

References

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