Two-center interference and nuclear motion effects in molecular harmonic generation

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Synopsis The momentum spread of the returning electron in molecular high-harmonic generation (HHG) influences the harmonic phase near the two-center interference minimum [1]. We investigate this effect using the strong-field approximation. Furthermore, we study effects of nuclear motion by simulating HHG in one-dimensional vibrating H_2 molecules irradiated by laser pulses with different wavelengths. The presence of two-center interference is confirmed. We show that the ratio of harmonic signals from the isotope pair H_2/D_2 can be satisfactorily reproduced by a simple semiclassical model [2]. The validity of the one-electron picture in the two wavelength regimes is established.

Atoms or molecules irradiated by intense laser fields undergo highly nonlinear processes that result in high-order harmonic generation (HHG) in the extreme ultraviolet (XUV) range. The physical picture is well explained by the three-step model [3]: laser-induced ionization takes place, followed by the active electron being accelerated by the laser field. In the final step, the electron returns to the atomic/molecular core where it recombines and releases an XUV photon. HHG from molecules is particularly interesting due to the presence of additional degrees of freedom compared to atoms: the multi-center nature, the nuclear vibration, or the molecular rotation.

In the usual picture of the HHG process, the harmonic frequency determines the momentum of the returning electron. Due to the multi-center nature of the molecule, under certain conditions, destructive interference takes place [1].

Around the minimum, the harmonic phase exhibits a jump, but this rapid variation has a certain width. We study the influence of the momentum spread of the returning electron on this width. Within the usual strong-field approximation model [4], the returning electron is described as a plane wave. The recombination matrix element is zero at the two-center interference minimum, implying a phase jump.

However, taking into account the momentum spread can explain the smoothness of the phase variation. The predicted width is bigger than that due to Coulombic effects in the case of a returning electron with fixed momentum.

We solve the time-dependent Schrödinger equation fully numerically for a one-dimensional vibrating H_2 molecule in a strong laser field. To emphasise the effects of the nuclear vibration, we calculate the ratio of the harmonic signals for the D_2/H_2 isotope pair. We employ 800 nm and 1500 laser wavelength.

The ratio clearly shows the two-center interference effect [1]. Moreover, the ratio is satisfactorily predicted at both wavelengths by an intuitive semiclassical model [2] based on classical trajectories and the vibrational wave-packet motion in the molecular ion.

We show that HHG at 800 nm is in essence a one-electron process. In contrast, for 1500 nm wavelength, the interaction of both electrons needs to be taken into account, the single-active electron picture becoming inadequate.

References

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