Quantum optical models of high-order harmonic generation in two-level system

PhD theses

Gombkötő Ákos

University of Szeged
Department of Theoretical Physics
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1. Introduction

High-order harmonic generation (HHG) is a strongly nonlinear optical phenomenon, during which material interacts with a high-intensity laser field, resulting in scattered radiation containing photons with energies that are multiples of the excitation photon-energy, up to a cutoff frequency. The phenomenon has been observed for the first time in 1987 using gas phase target, and it was soon realized that –assuming enough, phase-ordered harmonic– HHG can be utilized for the generation of short pulses (1). Since then, the study of this process has created literature of significant size. The focus of interest of the scientific community is primarily the possible realizations of higher efficiency and higher harmonic yield.

Today the HHG is one of the most important basic phenomena in attosecond physics, as this is the only, widely used method for the generation of short pulses beyond a limit. Therefore, it is unavoidable a part of experiments aiming to reach extreme temporal resolution of the dynamics of electrons in atomic, molecular, and solid-state targets. Attosecond physics is also decisive from the perspective of future research direction, and as finer temporal resolutions are becoming possible, the road to investigate newer phenomena is opening up.

The theoretical modelling of high-order harmonic generation, and the widely used, relevant concepts have been defined –for historical and computational reasons– overwhelmingly within the semiclassical approach. It is generally accepted that the process of HHG is practically determined by the interaction of electrons and the electromagnetic field. Accordingly, three elements can be separated in the models: the material system, the exciting field, and the scattered radiation field. Categorization of models can be done based on which of these three elements is considered to be explicitly quantized, and which are approximated classically.

In the traditional semiclassical models, both the excitation and the harmonics are present as classical objects, only the electrons are modelled quantum mechanically. Although such an approach can be generally considered successful in spectral calculations, it is inadequate for capturing the nonclassical, quantum optical aspects of the process.

However at present, when the role of photonics is getting more important in technological applications, it is desirable that basic phenomena get a detailed description in a formalism that can account for the quantized nature of the material system and of the electromagnetic field. Although earlier calculations involving the quantum optical description of high-intensity light-matter interaction exist since the early '80s [see review article (2)], the topic invited renewed interest. To mention a few recent results:
-It has been experimentally demonstrated, that photon statistics of the excitation is modified in such a way, that the generated high harmonic spectrum can be rendered (3).
-In molecular and atomic samples it has been demonstrated that sidebands, close to the even harmonics can be present, which are not necessarily present in semiclassical theories (4).
-Perturbative calculations imply that the quantum state describing the harmonic spectrum has modes with non-classical sub-Poissonian, or squeezed state (5).

Understanding relevant aspects of the process can contribute to the development of new sources of short optical pulses, in which nonclassical properties of light –such as squeezing, entanglement, sub-Poissonian photon-statistics– plays an important role.

2. Goals

Although earlier results exist regarding the quantum optical modelling of HHG, there are many open questions connected to whether and when the spectral and photon statistical properties differ significantly from the usual phenomenological descriptions. My goal was to make the first step on making a detailed exploration of this problem. In order to do this, I strictly limited my investigation to the fundamental, microscopic description, neglecting the macroscopically important wave-propagation and interference phenomena as well as the relaxation processes.

The most important investigated question was about what predictions models involving combinations of the classical and quantum description of the excitation and scattered radiation are capable of. Specifically what classically not treatable consequences the process of HHG may have. Due to the novelty of the topic, even a qualitatively correct description can be of use, therefore instead of precise calculations corresponding to realistic systems, I incorporated the material properties only as simplified parameters. To make the discussion transparent, I choose the material system to be a two-level atom, well known in the literature of quantum optics.

3. Applied methods

The model of interaction between laser fields and bulk solids (in one-electron picture, using dipole-approximation and velocity-gauge) can be reduced to the interaction of independent N-level systems with the electromagnetic field. In my work, I used the following assumptions:
-The excitation field before interaction can be characterized by coherent quantum state.
-The dipole approximation and the independent-particle approximation is valid.
-The intensity is of moderate magnitude, the two-band approximation is acceptable.

With these assumptions, I reduced the problem to the dynamics of independent two-level systems. In order to make the physical processes more transparent, I focused on the (numerical and analytical) treatment of the special case when the material system is a single two-level "atom".
It is worth noting that the semiclassical model of a two-level system can reproduce most qualitative features of the experimentally measured spectra, that is, the presence of a plateau with a cutoff; and (within moderate intensities) the linear dependence of the cutoff frequency on the peak field-strength (6-7).

In my work, I treated separately and compared properties of classical excitation induced quantized harmonic radiation, and quantized excitation induced classical radiation.

I gave special attention to the special case of monochromatic excitations, as it is more available for analytical investigation, and I determined the parameter-dependence of the dynamics. The inferences are identical for the case of pulsed excitations with rectangular envelopes.

For the case of monochromatic excitations, I derived analytical results that allows a simple evaluation of the spectral structure for given parameters. I gave the parameter $\delta\omega$ (that is, the detuning of the two spectral lines near the even-order multiple of the base harmonic) analytically.
I note that in the dissertation, I called these optical lines even-order harmonics for the sake of simplicity, but in the literature, it is primarily (but not exclusively) referred to as "Hyper-Raman lines".

It turns out that the value of $\delta\omega$ strongly influences the quantum optical properties of spectral lines. In the dissertation, I gave special attention to the case of parameters that maximizes $|\delta\omega|$, that is, when the two lines belonging to even harmonics have the highest spectral separation.

In order to incorporate the quantized nature of the excitation into the model, I worked out a phase-space method, based on Von Neumann's idea. The calculational scheme rests on using the Neumann coherent lattice basis, which allows the simple evaluation of the time-dependent Wigner-function characterizing the excitation {II}.

My numerical calculations show that the backaction on the excitation modes (which can be significant depending on the choice of parameters) has limited effect on the dipole-operator expectation value. In a formalism, in which the excitation is quantized but the harmonic radiation is calculated classically –that is, where the time-dependence of the dipole-operator expectation value is the source of harmonic radiation– I identified the lowest-order term causing spectral modification. For short interaction times, this modification is negligible.

I set up an effective model, in which the excitation is classical, while the scattered radiation is incorporated as quantized object {I}. Within this model, I developed an approximate calculational method for the dynamics of the collective quantized radiation field, based on the operator expectation values Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy. I calculated photon number expectation-values and quadrature-variances.

The photon-statistics, that is, the one-mode photon auto-correlation functions and the intermodal photon cross-correlation functions were calculated in one- and two-mode approximation numerically, giving an approximation analytically {IV}.

4. New scientific results

I. I identified the spectral structure of the harmonics induced by monochromatic excitation: it contains odd-order harmonics, and dual spectral lines near the even-order multiples of the base harmonic, which lines I called even harmonics in the dissertation. I calculated analytically the parameter dependence of $\delta\omega$, which characterizes the even harmonics.

I derived a (semiclassical) formalism, in which the first-order perturbation calculations is already in agreement with numerically calculated spectra within an acceptable relative error {IV}. For short interaction times, the incorporation of the quantized nature of the excitation in the model does not change the dynamics of the dipole operator significantly {II,III}.

II. The position of quantum optically calculated spectral lines coincide with the semiclassically calculated ones, and the classical three-step model can, at least qualitatively, give an acceptable explanation for the order of appearance of the harmonics {I,V}.

At the same time, the even harmonics calculated within quantum-optical model are often significantly more intense than the classical calculations would imply, and there is a difference in the dependence on initial conditions. I gave qualitative explanation analytically, through low-order perturbation calculation.

III. The harmonic radiation always contains modes characterized by squeezed quantum state (the odd harmonics are usually weakly squeezed), the number of which, as well as the degree of squeeze strongly depends on the excitation parameters. I made a detailed investigation for the case of monochromatic excitation. The choice of parameters fulfilling $\delta\omega = 0$ results in strongly squeezed even harmonic radiation, while the maximization of $\delta\omega$ results in anti-squeezed even harmonics {IV}.

IV. It generally stands that, (ignoring trivial oscillations) the odd-order harmonic modes are characterized by Poissonian photon-number distribution. From the quantum-optical perspective, we can assign interesting properties to the even harmonic lines, which, depending on the parameters and the interaction time may be of sub-, or super-Poissonian photon-statistics {I,IV}.

Of special interest is –concerning the generation of harmonics with non-classical quantum state– the case of monochromatic excitation, with parameters locally maximalizing $|\delta\omega|$. In such cases, the quantum state of the even harmonic modes spans only the vacuum- and one-photon states in Fock-space.
It is important to note that the other special case of $|\delta\omega| = 0$ corresponds to the generation of even harmonics with initially sub-Poissonian statistics, that grows to super-Poissonian.

V. The correlations between odd harmonic photons are, using the standard definition of correlation, practically always of unit value, independently the excitation and interaction parameters. Similarly, there is significant anti-correlation between odd and even harmonic photons in all case studies by me.
However the cross-correlation between even harmonic photons can be, depending on the parameters and the type of excitation either over unity, or nearly zero.

In the case of monochromatic excitation, if $|\delta\omega| = 0$, the subset of the field containing the even-order harmonics are characterized by strong mode-mode cross-correlations, while the modes individually are characterized by squeezed states. For the case of maximalized $|\delta\omega|$ even harmonic subset is characterized by strong anti-correlation, while the modes are individually in a strongly non-classical state. My analytical calculation shows that the anticorrelations correspond to nonclassical entanglements {IV}.

In summary:
-Within a semiclassical model I expressed the dependence of the spectral structure on excitation and interaction parameters, which primarily affect properties of the even harmonics.
-From a different approach (quantized excitation, classical harmonics) I concluded that the backaction on the quantum state of the excitation does not modify the harmonic spectra significantly.
-This justifies an effective model of HHG, in which the scattered radiation is quantized, while the excitation is assumed to be classical.
-Within our model, containing the fundamental, simplest material model, HHG can be a source of (depending on the parameters) squeezed or sub-Poissonian even harmonics.

Experimental verification of the predictions and potential application is possible in a similar experimental arrangement with which hyper-Raman lines were measured (4). Such experimental arrangement can also function as a source of broadband source of non-classical light.

5. Publications related to the theses

{I} Gombkötő Ákos, Czirják Attila, Varró Sándor, Földi Péter:
Quantum-optical model for the dynamics of high-order-harmonic generation, (2016)
Phys. Rev. A **94**, 013853
MTMT: 3099077

{II} Gombkötő Ákos, Varró Sándor, Mati Péter, Földi Péter:
High-order harmonic generation as induced by a quantized field: Phase-space picture, (2020)
Phys. Rev. A **101**, 013418
MTMT: 31142304

{III} Földi Péter, Magashegyi István, Varró Sándor, Gombkötő Ákos:
Describing high-order harmonic generation using quantum optical models (2021)
Photonics **8**(7), 263
MTMT: 32119823

{IV} Gombkötő Ákos, Földi Péter, Varró Sándor:
Quantum-optical description of photon statistics and cross correlations in high-order harmonic generation (2021)
Phys. Rev. A **104**, 033703
MTMT: 32210288

{V} Gombkötő Ákos, Varró Sándor, Keresztes Zoltán, Gábor Bence, Földi Péter:
A magasfelharmonikus-keltés kvantumoptikai vonatkozásai, (2021)
Szimpózium a hazai kvantumelektronikai kutatások eredményeiről, **pp. 64-68.**
MTMT: 32024350

6. Other publications

(1) Gy. Farkas and Cs. Tóth:
Proposal for attosecond light pulse generation using laser induced multiple-harmonic conversion processes in rare gases (1992)
Phys. Lett. A **168**, 447

(2) S. Varró:
Quantum Optical Aspects of High-Harmonic Generation (2021)
Photonics **8**(7), 269

(3) N. Tsatrafyllis, I. K. Kominis, Ivan Gonoskov, Paris Tzallas:
High-order harmonics measured by the photon statistics of the infrared driving-field exiting the atomic medium (2017)
Nature Communications **8**(1):15170

(4) E. Bloch, B. Samuel, D. Dominique, P. Stéphane, L. François, A. Magunov, M. Yann, S. Vasily:
Hyper-Raman Lines Emission Concomitant With High-Order Harmonic Generation (2019)
New Journal of Physics **21**(7)

(5) A. Gorlach, O. Neufeld, N. Rivera, O. Cohen, I. Kaminer:
The quantum-optical nature of high harmonic generation (2020)
Nature Communications **11**(1):4598

(6) You, Y., Yin, Y., Wu, Y. et al.:
High-harmonic generation in amorphous solids (2017)
Nat. Comm. **8**, 724

(7) G. Ndabashimiye, S. Ghimire, M. Wu, D. Browne, K. Schafer, M. Gaarde, D. Reis:
Solid-state harmonics beyond the atomic limit (2016)
Nature, vol. **534**, 520-523

(8) Á. Gombkötő:
Quantum optical properties of degenerate hyper-Raman lines appearing in high harmonic generation emitted by two-level systems
(In preparation)

(9) Á. Gombkötő, S. Varró, P. Mati, P. Földi:
Kvantált elektromágneses térrel keltett felharmonikusok,
Fizikai szemle **2020/6**

(10) P. Földi:
A magas felharmonikusok keltésének kvantumoptikai leírása,
Fizikai szemle **2017/10**

(11) Á. Gombkötő:
Solutions of the Ortvay Rudolf International Competition in Physics: Exploding Refrigerator (2016/10 Problem) (2021)
The Physics Educator Vol. **03**, 2120002