Three-photon excitation of quantum two-level systems

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Abstract: We demonstrate that a two-level system, in form of an InGaN quantum dot, can only be efficiency excited using an odd number of photons (1 or 3) while resonant two-photon excitation is strongly suppressed. © 2023 The Author(s)

1. Introduction

One of the most fundamental consequences of the quantum mechanical description of light-matter interaction are selection rules that classify transitions between levels. These multi-photon selection rules for transitions between two quantum levels depend on the symmetry of the states involved, the polarization of the light and the electric field approximation used to model the light-matter interaction Hamiltonian. If the excitation scheme involves N-photon of the same energy and polarzation, and the interaction Hamiltonian is considered in the dipole approximation, it can be shown that all even (odd) resonances are enhanced, based on the parity of the ground and excited states, while all odd (even) resonances are suppressed. This results from the fact that the electric dipole moment is an odd operator, coupling only wavefunctions with different parities. In this contribution, we investigate this fundamental prediction using InGaN quantum dots. Specifically, we demonstrate that these semiconductor quantum dots can be excited efficiently in a resonant three-photon process, whilst resonant two-photon excitation is strongly suppressed. In addition, we use the three-photon excitation process to investigate quantum-optical properties of the InGaN quantum dots.

2. Results

For probing this fundamental prediction of quantum optics, InGaN quantum dots are an excellent choice. Due to their emission wavelength in the range 450-500nm, also the wavelength of three-photon excitation of 1350-1500nm is conveniently accessible. Moreover, they don’t exhibit additional states in the bandgap that would result in a detuned effective two-photon excitation overlapping with resonant multi-photon excitation. An example measurement showing the photoluminescence intensity of a single InGaN quantum dot as a function of the excitation energy is presented in fig. 1. Resonant three-photon excitation is clearly observed for an excitation energy of 0.86 eV, while resonant two-photon excitation at an excitation energy of 1.3eV is suppressed. For higher excitation energies non-resonant two-photon absorption of the continuum of states of the wetting layer is observed as a step-like function in the spectrum around 1.35eV. Time-dependent Floquet theory is used to quantify the strength of the multi-photon processes and model the experimental results. The efficiency of these transitions can be drawn directly from parity considerations in the electron and hole wavefunctions.

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Fig. 1. Photoluminescence-excitation spectroscopy of InGaN quantum dots showing resonant three-photon excitation, while resonant two-photon excitation is suppressed. For higher excitation energies a non-resonant two-photon absorption into the continuum of states of the wetting layer is observed.

In addition, we exploit the three-photon excitation process to probe intrinsic properties of the InGaN quantum dots. In contrast to non-resonant excitation, slow relaxation of charge carriers is avoided which allows us to directly measure the radiative lifetime of the lowest energy exciton states. Since the emission energy is detuned far from the resonant driving laser field, polarization filtering is not required and emission with a larger degree of linear polarization is observed compared to non-resonant excitation.

3. References

[1] V. Villafane et al., arXiv:2202.02034 (2022)