Multipolar Transitions in Quantum Emitters: Beyond the Electric Dipole Approximation

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Abstract

In this presentation, we will investigate the multipolar optical transitions in quantum emitters, including rare earth ions, semiconductor quantum dots, and organic molecules. We will examine the electric dipole approximation commonly used to describe light-matter interactions and discuss naturally occurring systems that exhibit higher-order magnetic dipole and electric quadrupole emission. Then, we will illustrate how these quantum transitions can provide both a new way to probe magnetic light-matter interactions and a new degree of design freedom for active photonic devices. Specifically, we will demonstrate how the different symmetries of multipolar transitions can be exploited to identify, quantify, and control light emission, even at sub-lifetime scales.

1. Introduction

Pioneering research in metamaterials and nanophotonics has shown how the macroscopic electric and magnetic resonances of subwavelength nanostructures can provide unprecedented control over electromagnetic radiation. For example, there have been tremendous advances made in the nano-optics community by leveraging the electric dipole resonances of metal nanoparticles to serve as resonant optical antennas. At the same time, metamaterials researchers have realized fundamentally new material properties and phenomena by engineering the magnetic resonances supported, for example, by oscillating current loops in split ring resonators and fishnet structures.

However, in this talk, we will focus on a slightly different length scale, and discuss the microscopic electric and magnetic transitions in quantum emitters. When we talk about microscopic light-matter interactions, we often limit discussion to electric dipole transitions. Within the dipole approximation, the electrons in an atom or molecule experience electromagnetic waves as a time-varying but spatially uniform field. The local oscillating electric field produces a linear force, which can mediate an electric dipole transition such as the example depicted in Fig. 1(a) between a spherically symmetric *1s*-orbital to an anti-symmetric $2p_z$-orbital. And in reverse, the transition from $2p_z$ to *1s* can produce a local time-varying current that radiates like an infinitesimal electric dipole source.

Although often neglected at optical frequencies, there are also higher-order transitions such as magnetic dipoles [1]. Magnetic dipole transitions are mediated by the Lorentz force from time-varying magnetic fields, which produce a torque and therefore can facilitate rotational transitions. Fig. 1(b) depicts the magnetic dipole transition from a $2p_z$ orbital to a $2p_x$ orbital. Such transitions produce oscillating current loops that radiate like infinitesimal magnetic dipole sources.

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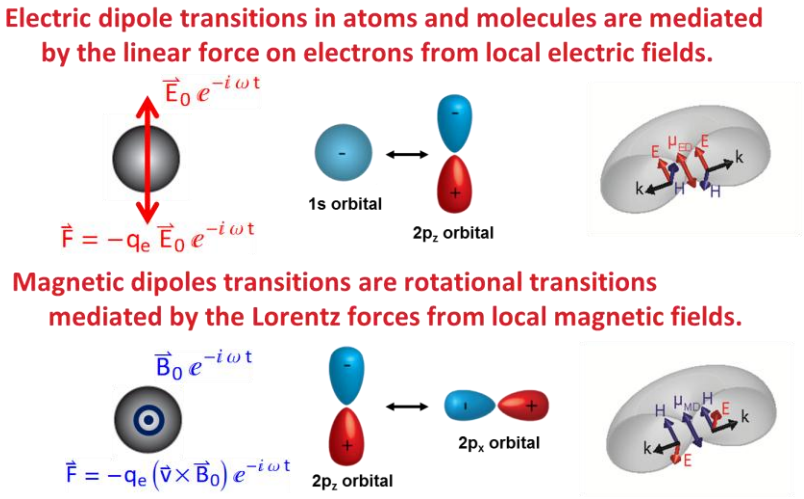

Fig. 1: Examples of the electric and magnetic dipole transitions in quantum emitters.

By comparing the dipole moments in the Bohr model, the oscillator strength of magnetic dipole transitions are often approximated to be five orders of magnitude less than those of electric dipole transitions. Thus, the microscopic origin of optical phenomena is almost always ascribed to electric dipole transitions. The goal for this talk will be to challenge this assumption by highlighting the existence and importance of higher-order optical transitions.

2. Exploiting the Symmetry Differences of Multipolar Transitions

Here, we will demonstrate how the different symmetries of multipolar transitions can be exploited to identify, quantify, and control light emission, even at sub-lifetime scales. Despite similar radiation patterns, magnetic and electric transitions have different symmetries with respect to polarization and phase as shown in Fig. 1. Thus, in an inhomogeneous environment, we can tailor interference effects and the local density of optical states to selectively enhance either electric or magnetic emission [2,3].

First, we will present quantum mechanical calculations to identify all magnetic dipole emission lines in the trivalent lanthanide series [4]. While many magnetic dipole absorption lines have been previously identified, only a few magnetic dipole emission lines are widely known and exploited. Such transitions offer new tools with which to study optical magnetic fields in metamaterials and related nanostructures. Using a detailed free-ion Hamiltonian, we show that there are many strong magnetic dipole emission lines throughout the visible and near infrared spectrum. These calculations also highlight the importance of magnetic dipole emission on mixed transitions, such as the 1550nm line in trivalent erbium (Er^{3+}) widely used in optical fiber amplifiers.

Next, we will present a novel spectroscopy technique to directly quantify the multipolar contributions from any mixed transition [5]. For example, based on self-interference effects near dielectric interfaces, electric and magnetic dipole transitions may be readily identified from their angular emission patterns. Using a microscope-coupled imaging spectrograph, we have experimentally measured the spectrum of light emission in both momentum and energy space. These measurements have allowed us to directly quantify the intrinsic spontaneous emission rates at each wavelength, and to directly probe the electric and magnetic local density of optical states.

Then, using the spectrally-distinct electric and magnetic transitions of trivalent europium (Eu^{3+}), we illustrate how self-interference can be used to modulate emission spectra at sub-lifetime scales [6]. By scanning a simple metal surface near a Eu^{3+} doped thin film, we demonstrate dynamic spectral tuning from 580-720nm without the need for a high-quality optical cavity. We will also present new device designs for directly-modulated on-chip optical sources that could enable high-speed modulation of emission polarization at sub-lifetime-limited time scales for optical communication.

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3. Conclusion

Multipolar transitions in quantum emitters clearly share many similarities, especially in pure electromagnetic terms, with the classical multipole resonances in metamaterial structures. Yet, there are also important differences between the microscopic transitions of quantum states and the macroscopic resonances of classical scattering objects. These similarities and differences suggest a variety of ways in which these two systems—one electronic, the other electromagnetic—may be combined, differentiated, and integrated.

The study of optical metamaterials and multipolar transitions can certainly complement one another. For example, advanced metamaterial structures provide the ideal electromagnetic environment in which to modify selection rules and enhance higher-order optical transitions, while light emission from such quantum transitions can be used to directly probe local electric and magnetic fields. The experimental techniques developed to quantify and exploit microscopic transitions may also provide new tools for the design and characterization of metamaterial structures and nanophotonic devices.

In addition to complementing one another, the investigation of metamaterials and quantum transitions can also enhance our understanding of microscopic light-matter interactions in unexpected ways. If time permits, we will show how the techniques developed to quantify multipolar transitions can also enable the study of more complex electronic systems including organic molecules, inorganic monolayers, and semiconductor quantum dots. Specifically, we will present experiments that isolate luminescent signatures from oriented excitons in layered nanomaterials and thus enable direct study of intra- and interlayer excitons species with distinct spectra and dynamics [7].

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