Nonlinear optical properties of 2D materials: from graphene to silicene and to other 2D materials

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Nonlinear optical (NLO) materials are the basis for several photonic and optoelectronic devices and applications. Among them, the 2D materials have attracted enormous interest due to their outstanding optoelectronic properties which endow them with great possibilities for a plethora of applications, as e.g. in ultrafast lasers, optical information processing and storage, telecommunications, sensors, etc.

The pioneer of 2D materials, graphene, being characterized by chemical inertness, zero bandgap and exhibiting dispersibility problems, is of limited use in photonics and optoelectronics. However, some graphene derivatives, such as: graphene oxides (GOs), graphene acid, fluorographene, nitrogen- and/or boron-doped graphenes and others, being readily dispersible in aqueous and/or other common organic solvents and exhibiting non-zero energy bandgap, seem to have great future and have gained increasing interest as they allow the efficient tailoring of their NLO response towards NLO materials with custom-made properties.

The popularity of graphene has triggered the interest for other graphene-like 2D materials based on other IVa group elements (Si, Ge, Sn, Pb), which share some of the outstanding properties envisaged for graphene. Recently, silicon nanosheets (SiNSs) have revealed comparable and even larger NLO response than graphenes, emphasizing their potential for 2D-material-based photonics and optoelectronic applications and devices.

During last years, 2D materials of the type MX_2, with M a transition-metal atom (Mo, W, etc.) and X a chalcogen atom (S, Se, or Te), also known as transition metal dichalcogenides (TMDCs), have also triggered the research interest, for their potential for micro-electronics and nano-photonics applications. Layered TMDCs, depending on their structural phases (trigonal prismatic-2H or octahedral-1T) exhibit semiconducting or metallic properties. The diversity of crystalline structure and structural phase of the d electrons, as well as the number and type of layer stacking sequences of TMDCs, result in a broad range of opto-electronic properties of these van der Vaals nanostructures. Consequently, it is reasonably expected that the engineering of crystalline structure of TMDCs can modify their NLO response as well.

Another class of 2D materials, with very interesting optoelectronic properties are some atomically thin non-van-der Waals (vdW)-layered nanostructures. Among them, hematene and magnetene, the two archetypical 2D iron-ore magnetic materials presenting thicknesses down to the atomically thin sheets have very recently attracted the research interest.

In this presentation, we will review some recent experimental findings of our group pertaining to the nonlinear optical properties of these 2D nanostructures, including graphene derivatives, silicon nanosheets, some vdW-layered TMDCs and some non-vdW-layered iron oxides. The NLO response of these nanostructures was investigated in time scales from ns to fs, using Z-scan and pump-probe Optical Kerr effect (OKE) techniques. In addition, the dynamics and the operating physical mechanisms responsible for the NLO response will be discussed. Additionally, the critical role of defect-engineering, chemical functionalization, size-effects, and crystalline phase on the NLO response of these 2D materials will be highlighted.

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