Intersubband plasmons induced negative refraction at mid-IR frequency in heterostructured semiconductor metamaterials

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Abstract. In this contribution, we report on negative refraction effect occurring in layered semiconductors. The origin of this effect is attributed to the presence of intersubband plasmon resonance induced by the electronic confinement of the electrons in thick quantum wells. Relying on the effective medium theory, we analyse both the in plane and the out of plane effective optical properties of highly doped ZnO/ZnMgO semiconductor material. We theoretically show and experimentally demonstrate that thicknesses and doping levels of each layer can be carefully chosen to feature strong intersubband transitions, leading to type 1 and type 2 hyperbolic response. Understanding the optical properties of intersubband materials in the frame of hyperbolic materials would shed new light on which physical mechanisms are controlling the radiative decay of intersubband plasmon excitations. This approach could be further utilized to designing efficient mid-IR sources.

1. Introduction
Metamaterials are artificial materials that feature uncommon physical properties. In optics the field of metamaterials has received tremendous interest over the last few years, with the demonstration of various unexpected and intriguing effects such as ultra-high refractive index and extraordinary optical activity. However optical metamaterials are mainly known owing to the fascinating possibility of realizing devices based on negative index of refraction, also called negative index materials (NIM). A great deal of attention on this domain has been attracted due to the potential of metamaterials in revolutionizing both fundamental and applicative research. Material with negative index of refraction was firstly been considered in a theoretical paper by Veselago in 1968. In this latter paper, it is shown that negative index can be obtained in a material presenting simultaneous negative dielectric permittivity and magnetic permeability, in brief by adjusting two and matching electric and magnetic resonances in the material. It implies that in such systems the absorption is quite relevant. In order to mitigate optical losses, lots of efforts have been made to reduce the absorption losses, considering for example gain material. A more efficient approach has been proposed in recent years. It consists on exploiting hyperbolic dispersion, i.e. opposite signs of permittivity along orthogonal in- and out-of plane directions. Affecting the optical response only along one direction, hyperbolic dispersion can be realized in anisotropic medium using a single optical resonance. In such a system, also dubbed hyperbolic metamaterial (HMM), one of the electric or magnetic resonance is replaced by leveraging on the material anisotropy. The conceptual and experimental simplification also comes with a drastic reduction of the optical losses. The main idea behind HMM is to make use of an alternating composition of both resonant and non-resonant materials. Peculiar attention has been given to semiconductor HMM [1-3] because of their functional and fabrication advantages, substituting the metallic resonant part by a highly doped semiconductor.
In this paper we present theoretical and experimental results confirming negative refraction in highly doped layered semiconductor materials and highlight the fact that this behaviour is essentially driven by the intersubband transitions (ISBT) in the quantum wells (QW) [4].

1 – Samples: materials and geometry
The physical system proposed in this paper is a stack of alternated layer of doped ZnO and undoped MgZnO forming a quantum wells along z direction grown on native ZnO substrate to reduce the dislocation density for sharper ISBT. The choice of ZnO is mainly related to the possibility of reaching very high doping level and because of its non-polar growth possibility, therefore avoiding the detrimental quantum confined Stark effect [5]. A scheme of the system is reported in Fig. 1a.

2 – Model
The problem was mathematically treated following Maxwell-Garnett theory, in the presence of both the QW and the barrier to obtain realistic material permittivity. Details on the model to account for phonon contribution, and anisotropic ISBT response will be discussed during the presentation. The main results are summarized in Fig. 2a and 2b that show both the in- and out-of-plane components of the real part of the effective medium permittivity. Fig. 2c summarizes the permittivity signs: the optical properties range from pure metal to pure dielectric passing through HMM type 1 and 2. We show that T1 HMM originates from the intersubband plasmon response of the material.

3 – Experimental results
The theoretical prediction were tested experimentally with two samples of respectively 35 and 4 nm thickness ZnO quantum wells. According to the figure 2, the former should exhibits negative refraction while the latter should behave as a conventional dielectric material in the 1400-1800 cm\(^{-1}\) wavenumber range. The samples are composed of 15 couples of QW-barrier having total thicknesses equal to 590 and 125 nm respectively.

Negative refraction have been detected by using a blade to stop either the positive refraction or the negative refraction depending on the sample orientation (Fig. 3a). According to the figure in presence
of negative refraction a maximum in transmission for θ<0 is expected since the positive refraction is stopped. Symmetrically, a minimum is expected for θ>0. This result is confirmed by experimental spectra reported in Fig. 3b.

Figure 3. (a,b) Scheme of the experimental setup: stopping the positive and negative refracted laser beam respectively. (c) Experimental results: transmission ratio between half-blocked and unblocked beam. It shows a convexity inversion around 2000 cm$^{-1}$ with the inclination angle θ according with the theoretical model.

4 – Conclusion
In conclusion we have experimentally demonstrated the possibility of reaching negative refraction due to intersubband transition in a layered semiconductor system. The experimental results are in agreement with the theoretical model. Although it is clear that intersubband designs are composed of several subwavelength layer of Drude-like materials, this work connects for the first time the concepts of intersubband plasmons with the photonic response of hyperbolic metamaterials. Due to the advanced photonic properties of HMM, we believe that these results could initiate new design for efficient mid-IR light emitting sources.

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