Surface plasmon lasers

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Chapter 1

Introduction

1.1 Wave confinement and surface plasmons

Confining a wave of wavelength λ to a limited volume of space with typical dimension L is quite an easy task provided that the dimension L is larger than the wavelength λ. Musical instruments and lasers provide prime examples of this idea. In both cases the confinement gives rise to resonant enhancement of the wave-field amplitude. In contrast, when the volume is much smaller than the wavelength, the wave cannot be confined in this resonator-like fashion and resonant enhancement is non-existent.

Nature, however, does provide a totally different system of wave confinement, namely on an interface between two materials. There, surface waves can exist and the name betrays the nature of the wave phenomenon: it is confined to the interface. Ocean waves and coastal edge waves [1, 2] are prime examples of this particular wave phenomenon, and so are surface plasmons (SPs). The latter are electromagnetic-like waves that hug the interface between good metals, such as silver and gold, and a dielectric.

Surface plasmons consist of light coupled to free electrons on a metal-dielectric surface and hence are strongly confined to this surface. The electromagnetic field induces a temporal charge redistribution in the metal and it oscillates the electrons at optical frequencies. The strong confinement leads to a large enhancement of the wave amplitude, opening up the possibility of strong light-matter interaction.

During the last 20 years the study of surface plasmons has experienced an enormous revival, mostly as a consequence of novel and advanced nanofabrication techniques. It has led to a large variety of applications of surface plasmons, such as sensors based on surface-enhanced raman-spectroscopy of molecules [3]. Here, SPs are employed to increase the light-matter interaction and drastically enhance the single-molecule signal up to 10-orders of magnitude [4]. Closer related to the work in this thesis are optical metamaterials [5, 6], where artificial building blocks are used to create materials with unprecedented optical properties such as a negative refractive index [7–9]. These metamaterials have been used to create ultra-thin lenses [10–13], waveplates [14–16], and rudimentary invisibility cloaks [17]. A particular example of a simple metamaterial in which SPs play an important role is a metal hole array. It consists of a metal film perforated by a lattice of sub-wavelength nano-holes. Metal hole arrays form two-dimensional crystals for SPs. They exhibit extraordinary transmission [18], meaning that more light is transmitted than is expected from the surface area of the holes. This extraordinary transmission is mediated by SPs.

Absorption in the metal poses a limitation on the application of SPs. This absorption is caused by electron scattering (Ohmic loss) and hence unavoidable. To mitigate this absorption, an optical gain material next to the metal surface can be employed for loss compensation [19]. Due to the strong confinement of the light, only a thin ($\sim$ 120 nm) gain layer is needed: SPs with an energy equivalent to photons with a free space wavelength of 1500 nm are confined within 200 nm from the gold-semiconductor interface. Figure 1.1 shows a schematic of the layer stack of our samples, consisting of a 100 nm thick gold layer on a semiconductor (InGaAs) gain layer top of a InP substrate. It also illustrates the confinement of the SP field at the Au-semiconductor surface. As soon as the Ohmic loss and all other losses of the SP mode are compensated, SP-laser action can occur [20].

Lasers are known to emit coherent, monochromatic, and strongly directional beams. There are two essential components to a laser: a (pumped) gain medium and a resonator. The resonator confines the laser mode and supplies the feedback needed to obtain coherence. The aim of this thesis is to understand SP lasers and to investigate to which extent they can be described by traditional laser theory. We focus primarily on the resonators which, in our case, are formed by metal hole arrays.

1.2 Surface plasmon lasers

SP lasing has been observed in several resonator geometries, from nano particles to metal hole arrays. The first claim of SP-laser action was based on observations of isolated 14 nm-large nano particles [21]. However, these results are disputed on theoretical and experimental grounds and have not been reproduced by other groups to date. Next, Hill et al. [22] demonstrated SP lasing in metal-coated semiconductor nano pillars, in which a localized resonance in such a pillar forms a zero-dimensional resonator. SP-laser action also has been observed in nano-wire systems, in which the gap between a

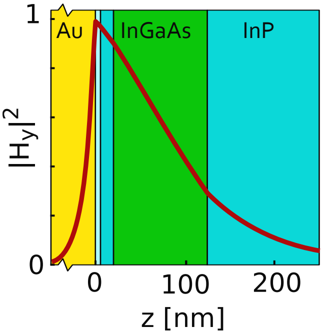

Figure 1.1: A side-view of our devices, consisting of an Au metal hole array on top of a semiconductor (InGaAs) gain layer on a InP substrate. The red curve shows the intensity of the H_y component of the SP-field. The SP field decays away from the metal-dielectric interface. Details are given in Fig. 2.1.

semiconductor nano wire and a silver surface serves as a one-dimensional resonator [23]. These experiments were later extended to two-dimensional geometries, where the feedback is provided by total internal reflection of the SPs in the semiconductor gain medium [24].

Two-dimensional resonators can also be based on distributed feedback, where the optical feedback is not provided by a Fabry-Pérot cavity comprising two highly reflective mirrors but by scattering on a periodic array, either in the form of holes in a metal [20, 25] or metal particles on a substrate [26–30]. Both periodic arrays support SPs and SP lasing, and they form two-dimensional crystals for SPs. For metal hole array SP lasers, the resonator is formed by the reflection of traveling SPs on the holes, as discussed below. In contrast, for particle-array SP lasers, the interplay of localized particle-resonances and non-localized lattice resonances typically plays an important role and the feedback can be described by coupled localized harmonic oscillators. Research on particle arrays has demonstrated, among others, lasing in the strong-coupling regime [30] and the influence of randomness [29].

SP lasing in metal hole arrays has been demonstrated at wavelengths ranging from the visible regime (∼ 0.6 µm) to the THz regime (∼ 100 µm). The first demonstration originates from our group in Leiden, for SP lasers operating at telecom wavelengths (∼ 1.5 µm) using a solid-state semiconductor gain medium [20]. Later, others used molecular dyes as gain medium in the visible regime [25]. Metal hole arrays are also used as resonator for lasers

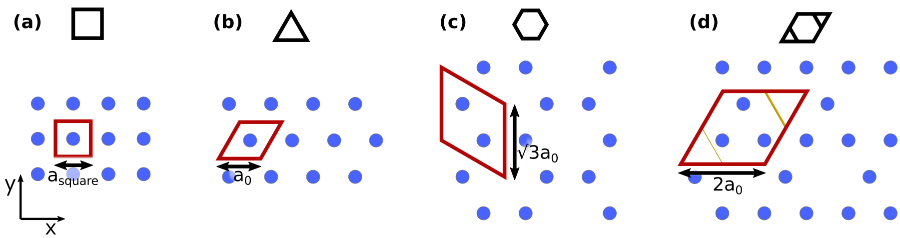

Figure 1.2: Schematic images of the 4 different two-dimensional crystal lattices studied in this thesis: (a) square, (b) hexagonal, (c) honeycomb and (d) kagome. Their unit cells are indicated by red parallelograms.

at much longer wavelengths, corresponding to THz frequencies [31, 32]. However, at these frequencies the confinement of SPs above the surface is weak and the confinement can only exist due to the specific structure on the surface; hence, these SP are called spoof-surface plasmons [33] to indicate that they differ considerably from ordinary SPs.

1.3 Crystals and band structures in two dimensions

Crystals are periodic structures that can be described by their unit cell and by their Bravais lattice or associated reciprocal lattice vectors $\vec{G}_i$. Besides a translation symmetry, most crystals exhibit additional mirror and rotation symmetries, which can be described by point groups. In this thesis we will consider two-dimensional crystals for SPs, consisting of lattices of metal holes in a gold film. Figure 1.2 displays a schematic overview of the studied crystals, which are square lattices (C_{4v}-point group) and three hexagonal-based lattices (C_{6v}-point group): hexagonal, honeycomb and kagome. The spacing between the holes is comparable to the wavelength of the SPs. The hexagonal-based lattices have the same symmetry, but increasingly more complex unit cells.

The dispersion relation describes the relation between the wavelength $\lambda = 2\pi/k$ (or wavevector $\vec{k}$) of a wave and its energy (or frequency ω). It plays a central role in solid-state physics, where it forms, among others, electronic conduction and valence bands, and determines the performance of diodes, LEDs and transistors. It also plays a central role in the description of optics and plasmonics in crystals. In both cases, crystals severely alter the dispersion relation.

Periodic structures and crystals scatter waves and create standing waves

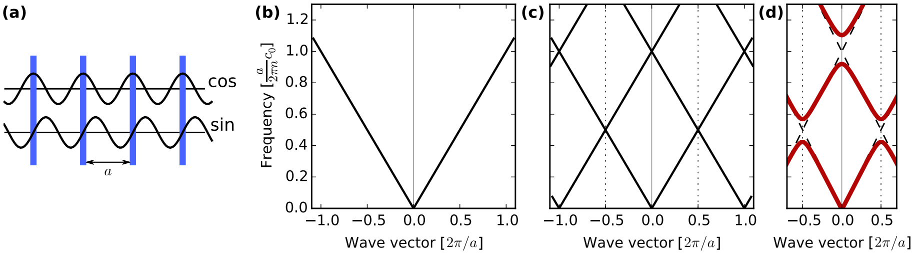

Figure 1.3: (a) One-dimensional crystal lattice with lattice spacing a. Dispersion of (b) free waves, (c) waves in a periodic lattice, and (d) waves in a scattering periodic lattice.

that can completely stop the wave propagation in specific crystal directions. The formation of such bands in the dispersion relation is due to the combination of scattering on the unit cell and their periodic nature. This is most easily explained for a one-dimensional infinitely-large crystal. Figure 1.3(a) shows several unit cells of a one-dimensional crystal with lattice spacing a and lattice vector $G = 2\pi/a$. Figure 1.3(b) shows the linear dispersion of a free wave. The slope is linked to the effective refractive (group) index via the relation $d\omega/dk = c/n_{eff}$. Figure 1.3(c) shows the dispersion of a wave in a one-dimensional crystal. The periodicity of the crystal induces a periodic repetition in the dispersion relation; wave vectors spaced with a lattice vector $\vec{k}_1 - \vec{k}_2 = \vec{G}_i$ are equivalent such that all information is contained in the first Brillouin zone $\left[-\vec{G}_i/2, \vec{G}_i/2\right]$. At higher order Γ-points ($k = 0$, $\omega > 0$) left- and right-traveling waves cross and their wavelength fits on the lattice. Figure 1.3(d) shows the influence of scattering on the holes in the unit cell. The scattering couples part of the left-traveling wave to the right-traveling wave (and vice-versa); standing waves are formed and avoided crossings appear. The anti-symmetric (sine-type) standing wave has nodes on the holes, while the symmetric (cosine-type) standing wave has anti-nodes on the holes, as shown in Fig. 1.3(a). Hence, these two standing waves have different energies $\omega_\pm = \omega_0 \pm \gamma$, where γ is the amplitude scattering rate, and a stop-gap is formed, i.e. an energy range in which no waves can travel in a certain direction.

In two dimensions, the band structure is more complex than in one dimension as waves and scattering in additional directions have to be included [34]. We study the formation of SP bands in two-dimensional crystals with square symmetry in chapters 2 and 3, and with hexagonal symmetry in

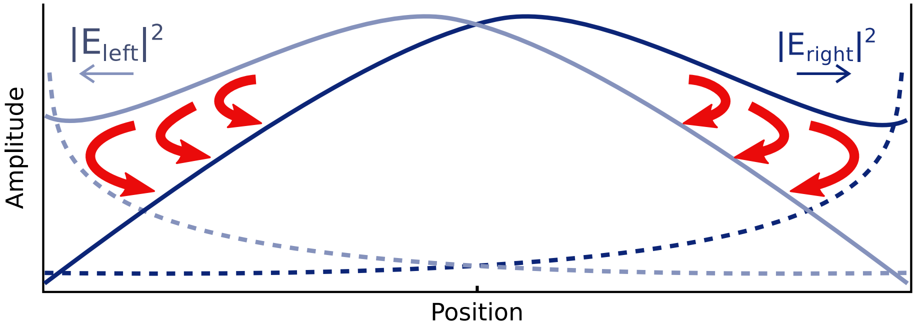

Figure 1.4: Fields inside an one-dimensional distributed feedback laser. The dashed curves show field profiles when no coupling is present, the solid curves show the profiles in the presence of coupling.

chapter 5. We also study the influence of the shape of the unit cell on the scattering rates and its link to the scattering by a single hole.

1.4 Lasing in finite size crystals

The analysis of the dispersion relation of two-dimensional crystals presented in the previous section works fine for very large crystals, but is insufficient for the description of SP-laser action in crystals of finite size. This finite size alters the band structure; it breaks apart the continuous band structure of infinite-large crystals into discrete modes. Suited for lasing in finite size crystals is distributed-feedback laser theory [35]. This theory describes the laser field as traveling waves in real space that are, again, coupled via scattering. Figure 1.4 shows how this scattering confines the field to the center of a one-dimensional device.

In two-dimensional crystals traveling waves in additional directions should be included in this distributed-feedback laser theory; it becomes more complicated and no analytical solutions are known. Numerical modeling of the combination of strongly-confined SPs and gain is challenging and hence experiments are invaluable in order to understand the behavior of such systems. The first realizations of lasing in two-dimensional crystals was in photonic crystals. Since then the field blossomed and produced, among others, Watt-class surface-emitting photonic-crystal lasers [36]. Utilization of this knowledge can accelerate the development of SP lasers and SP sensors.

1.5 Outline of this thesis

In this thesis, we study SP-laser action in metal hole arrays. We try to understand these systems and connect this understanding to existing knowledge about lasers. The figure below schematically displays the contents of this thesis. The two columns indicate that we have studied SP-lasers in metal hole arrays with different geometries: square lattices (left column) and hexagonal-based lattices (right column). The two rows indicate that we have studies these structures both below lasing threshold (top row) and above lasing threshold (bottom row). Below threshold we measured the dispersion of the SPs and extracted information on their loss and scattering rates. Above threshold, we observed SP-laser operation and retrieved information about intensity and phase profiles, polarization, optical feedback and spatial non-uniformities. All these experiments have contributed to our understanding of SP physics in metal hole arrays.