Plasmonic High Harmonic Generation

Brian Kamer

Abstract— **High harmonic generation arises from intense laser beams where odd harmonics of a fundamental frequency up to a cutoff threshold are generated. This cutoff nth multiple is intensity dependent and the ionization potential of an atomic system. Surface plasmon polaritons have been shown to amplify the incident electric field greater than 1000 times allowing the direct coupling of a laser oscillator without the need for amplification.**

I. INTRODUCTION

S urface plasmon polaritons (SPP) have been demonstrated to be a feasible technique for high harmonic generations. The recent advances in nano-construction have allowed scientists to create specific nano-geometries to create waveguides. These waveguides can then be created so that incident light is squeezed into sub-wavelength apertures and coupled to surface plasmons (SP) via a metal/dielectric interface. The confined fields experience extreme enhancements. Polaritons are photons that are couple to surface plasmon and propagate along the interface due to the interaction of the photons with the free electrons in a conductor.

High harmonic generation is enhanced when the SPP are confined in such a tight manner and the creation extreme UV light pulses are possible. HHG is created when incident electromagnetic fields exceed the ionization potential of an atomic system and the freed electrons are accelerated toward and collide with the nucleus.

II. BACKGROUND

A. Surface Plasmons

Electrons in a metal respond to external electric fields by migrating to the surface. Optical frequencies are typically longer than the time it takes the free electrons to rearrange themselves. This leads to the electrons to oscillate at optical frequencies and these same electrons can be modeled as plasma. When these oscillations are coherently they are described as surface plasma oscillations, also known as surface plasmons (SP). The limit to the above model is obviously the plasma frequency of the metal, the electric field for a TM plane wave incident on a metal,

$$\vec{E_1} = (E_{x1}, 0, E_{z1})\, e^{-i(k_{x1}\vec{x} + k_{z1}\vec{z})} \tag{1}$$
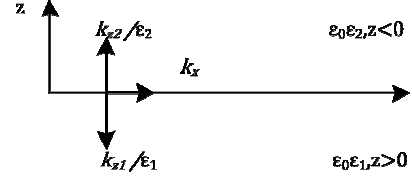$$\vec{E_2} = (E_{x2}, 0, E_{z2})\, e^{i(k_{x2}\vec{x} + k_{z2}\vec{z})}.$$

Fig.1 Wave vector directions.

From Maxwell's equations and the boundary conditions and $\nabla D = 0, \nabla H = 0$,

$$\frac{\partial \widehat{H}_{y1}}{\partial z} = \varepsilon_1 \vec{E}_{z1} \frac{\omega}{c} \tag{2}$$
$$\frac{\partial \widehat{H}_{y2}}{\partial z} = \varepsilon_2 \vec{E}_{z2} \frac{\omega}{c}$$

leads to

$$\varepsilon_1 \vec{E}_{z1} = \varepsilon_2 \vec{E}_{z2} \ \text{ and } \tag{3}$$
$$k_{x1} = k_{x2} = k_x.$$

The total wave vector is,

$$k_x^2 + k_{zi}^2 = \varepsilon_i \left(\frac{\omega}{c}\right)^2, \quad i = 1,2. \tag{4}$$

and solving for k_x,

$$k_x = \frac{\omega}{c}\left(\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\right)^{1/2}. \tag{5}$$

Taking the complex dielectric function $\varepsilon = \varepsilon' + i\varepsilon'$ and assuming ω and ε_2 is air, ε_1 is the metal,

$$k_x' = \frac{\omega}{c}\left(\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2}\right)^{1/2}, \tag{6}$$
$$k_x'' = \frac{\omega}{c}\left(\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2}\right)^{3/2} \frac{\varepsilon_1''}{2{\varepsilon_1'}^2}.$$

For k_x to be real, the signs of ε_1 and ε_2 must be opposite which is the case when the metal absorbs photons. The SP in (6) shows that as k_x increases, the dielectric function limits the frequencies of a SP and it lags the incident field
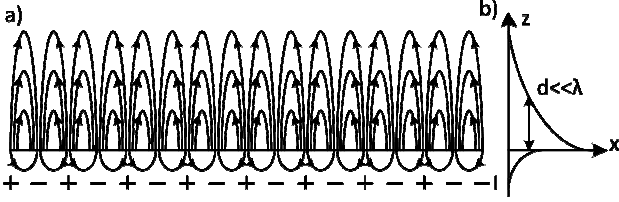
Fig.2 a) shows the electric field along the surface of the interface where the fields in the dielectric are in the positive z direction. b) Shows the field decay discrepancy between the metal and dielectric.

Fig.3 a) FDTD sim. Showing field enhancement b) Scanning electron microscope image of a v-groove [5]

frequencies. For example, red photons generate $k_{sp} \approx 1.03 k_0$ [1]. This momentum mismatch must be corrected in order for a photon to couple to the SP.

Surface plasmons propagate along the interface with the electric field component is perpendicular and decays exponentially. Also, no power radiates away from the surface, and SP takes on an evanescent field characteristic. However, the SP loses energy as it propagates due to being absorbed by the metal substrate. Solving the imaginary part of (6),

$$\frac{1}{k_x''} = \frac{c}{\omega}\left(\frac{\varepsilon_1' + \varepsilon_2}{\varepsilon_1'\varepsilon_2}\right)^{3/2}\frac{2\varepsilon_1'^2}{\varepsilon_1''}, \qquad (7)$$

$$L = \frac{1}{2k_x''} = \frac{c}{\omega}\left(\frac{\varepsilon_1' + \varepsilon_2}{\varepsilon_1'\varepsilon_2}\right)^{3/2}\frac{\varepsilon_1'^2}{\varepsilon_1''}.$$

where L is the propagation distance to the $1/e^2$ value.

There are three main techniques to conserve photon momentum. The first is to use a prism to increase the increase the photon momentum by $n \cdot k_x$. The incident angle is adjusted until there is evanescent coupling between the prism and the substrate. The next method is to use some sort of defect on the material surface, or to manufacture a hole, dimple or some other protrusion to change the period of the polariton. Lastly, a diffraction grating or manufacturing a periodic surface in the material that is to generate the plasmons can be used to generate polaritons.

B. High Harmonic Generation

High harmonic generation is a process that occurs when an incident laser beam is of sufficient intensity to photoionize an atom or when the electric field is on the order of,

$$\vec{E}_{atomic} = \frac{q}{4\pi\varepsilon_0 a_0^2}. \qquad (8)$$

$$a_0 = \frac{4\pi\varepsilon_0 \hbar^2}{m_e q^{\wedge}2}$$

where a_0 is the Bohr radius. The atomic field is on the order of $5.14 \times 10^{11} Vm^{-1}$. Incident irradiance of this magnitude is sufficient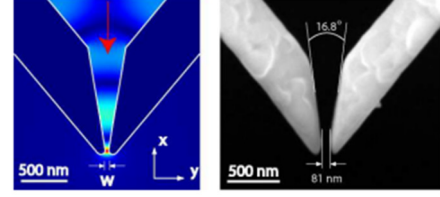 to ionize atoms, and these now free electrons move in the wake of the electric field. Depending on the polarization of the incident light, the electron motion in the plasma will either be circular or a very tight ellipse. Linearly polarized light gives the best probability of the freed electron recombining with its' nucleus. To achieve higher order harmonics the electron must accelerate much faster than the incident field incident field. This is accomplished with linearly polarized light and as the oscillating field changes direction, the electron is driven back towards the parent atom, is further accelerated until it collides with the nucleus. As a result of the collisional processes, only odd harmonics are generated. Analysis of the emission spectra displays the first few harmonics with the largest intensities which decay only for a few more octaves. Then the intensity plateaus for the majority of the octaves until some maximum frequency is achieved. The highest harmonic frequency that is generated is proportional to the incident irradiance, and is given by,

$$q_{max}\hbar\omega = 3.17K + I_p \qquad (9)$$

where K is the pondermotive energy and I_p is the ionization potential and is equal to (8).

C. Tapered Waveguides

Surface plasmons are not limited to the diffraction limit of light, and by coupling photons to the SP it was shown that it was possible to focus the incident wave below the diffraction limit. In [6] tapered metal v-grooves were constructed so that the SPP would propagate toward the narrow end of the grooves so that there was no cut of mode. The v-grooves are illustrated in fig.3. The authors created an array of v-grooves and placed a CCD in the far field and were able to record the output of the array. The field enhancement was calculated from the number of illuminated v-grooves, the far field intensity and the area of the v-groove apertures. The gain factor is approximately 10.

Successful generation of high harmonics with plasmon enhanced fields was demonstrated in [7]. The wave guide was designed using FDTD simulations and trial and error to obtain the dimensions that yields the largest field enhancement. One of the overall goals was to be able to modify off the shelf products in the construction of the waveguide, and a hollow cantilever for a scanning electron microscope was used. It the cavity is pyramid shaped and was initially filled with a thin layer of platinum before being filled completely with silver.

Next, the dimensions calculated from the FDTD simulation were etched out using a focused ion beam.

III. Surface plasmon enhanced HHG

The fabricated waveguide in [7] was placed in a glass cell filled with xenon gas to generate HHG. This cell was then placed in a vacuum chamber. The waveguide was pumped at 800 nm with 10 fs Ti:Sapphire laser with a 75 MHz repetition rate. The waveguide was directly from the oscillator with an irradiance that is below the ionization potential in (9). The waveguide has an elliptical aperture that is 2 μm x 4μm and 9μm deep and a 100nm opening with a 14° taper. The laser was focused down into to a spot size of about 5μm which is slightly larger than the opening.

With the laser being focused on the outer rim of the waveguide the beam is diverging when it enters the waveguide. The intensity of the field is give at $5 \times 10^{11} Vm^{-1}$ or about two orders of magnitude lower than the threshold for HHG. The diverging field is reflected and would be near grazing incidence forcing the condition in (3), and creating the conditions for SPP to form. The tapered cone is essentially a horn antenna in reverse, and as the SPP propagates to the tip it is below the cutoff frequency and is reflected when minor diameter of the waveguide wave half of the plasmon wavelength and generates a standing wave. This also leads to further enhancement of the electric field. The SP were initiated by the pump laser, a reflected approximately 400nm from the tip of the waveguide.

The emitted light also demonstrated the behavior of being confined beyond the diffraction limit. This arises from the fact that the SPP frequency is driven by the optical field, but the phase velocity is retarded by the slower moving SP and as a result the coupled photons. The electric field of the SP is perpendicular to the surface obeying,

$$n \times (\vec{E}_1 - \vec{E}_2) = 0 \qquad (10)$$

with the electric field folding and returning to the surface see fig(2). The cylindrical wave guide gives the normal component in the radial direction and the narrowing of the waveguide,

$$\vec{E} = -\nabla \Phi \qquad (12)$$

is maximized in the direction of propagation leading to maximum SPP enhanced field.

At the tip of the waveguide, the opening is 100 nm, and the beam divergence emitting from the tip,

$$\theta = \frac{\lambda}{\pi w}. \qquad (13)$$

The light emitted from the tip is not only UV but is extremely divergent and a transmission grating has to be used as glass is opaque to XUV. A photodetector was placed in the far field with a series of bandpass filters, and the detector was swept to measure the location of the mth diffraction order. Once the diffraction angle was known the nth harmonic was calculated from the Fraunhofer integral.

IV. Discussion

Typically, the output of a Ti:Sapphire oscillator is not sufficient to create HHG and the various methods of amplifying an oscillator results in the pulse repetition going down. SPP has been proven to be sufficient to amplify the optical fields to intensities large enough for high harmonic generation. Using this technique over the tradition method drastically reduces the complexity of the experiment by removing the amplifiers but it also maintains the pulse repetition rate of the oscillator. Another benefit of this method is that the HHG process takes place in a confined space where the incident fields are well above the ionization threshold. This eliminates the need for complicated phase matching techniques and by the nature of the sub wavelength aperture the pump beam and the first few harmonics are removed for the output signal. An added benefit of using silver for the creation of SP is that he has excellent thermal properties and inspection of the waveguide showed little or no signs of damage despite the terawatt level intensities.

V. Conclusion

It has been shown that surface plasmon polaritons can enhance optical fields to levels large enough for high harmonic generation. Furthermore, this technique eliminates much of the complexity required to amplify an oscillator as well as maintained the repetition frequency. It may soon be possible to carry out ultra-short pulse, extreme UV in the near field by illuminating the need for complicated techniques. Finally, using off the shelf components that can be readily modified makes this method a suitable candidate for the study of attosecond physics.

References

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