Nonlinear processes induced by the enhanced, evanescent field of surface plasmons excited by femtosecond laser pulses

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Abstract: Evanescent fields of surface plasmon polaritons (SPP) above metal surfaces can reach 1-2 orders of magnitude higher, nearly atomic field strengths in comparison to the relatively weak exciting laser fields of a femtosecond Ti:sapphire laser oscillator. We used these high plasmonic fields to study the characteristic SPP phenomena of intense field optics experimentally. It was found that both the intensity and the angular distribution of SPP emitted light depend nonlinearly on the exciting laser intensity in the higher-intensity, non-perturbative range of the interactions. These results are supported by our theory. At these strong excitations, an additional, depolarized, diffuse spectrum also appeared which can be attributed either to the fluorescence of Au, or to the non-equlibrium Planck radiation, originating from the fast cooling of the conduction electron cloud of Au excited by the femtosecond laser pulse.

OCIS Codes: 240.6680 Surface plasmons; 240.4350 Nonlinear optics at surfaces; 320.2250 Femtosecond phenomena

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1. Introduction

Surface plasmon polaritons (SPPs) are subject to extensive studies due to their unique properties, opening novel ways of studying the physics of the interaction of light with surfaces and surface electrons in the intensity range extending from perturbative phenomena up to extreme nonlinear optics. These studies, with their incentive approaches have lead to new results both in basic research (quantum electrodynamics (QED), intense field electrodynamics, attophysics [1-6]) and in applied nanosciences, opening up new perspectives in physical, chemical, biological studies [7,8].

According to very recent theoretical predictions [2-6], a quite new theoretical and experimental way is opened in which (in addition to the traditional works done at low excitation intensities) new investigations of intense field processes can be performed. The basis of this new possibility is the use of SPP-enhanced strong electromagnetic fields that can be produced even with a simple Ti:sapphire laser oscillator alone.

The aim of present work is the experimental study of fundamental physical processes in the high-intensity evanescent electromagnetic fields of SPPs in addition to the traditionally studied low-intensity phenomena [9]. The use of this evanescent field for different experiments is an interesting and challenging opportunity even in itself but it is also important for possible applications. Relatively low intensity femtosecond Ti:sapphire laser oscillators with ~10^{11} W/cm^2 peak intensity can create up to ~10^{14} W/cm^2 intensity with the help of the SPP evanescent fields [2,3,9-11]. Stepping into this high-field region, non-perturbative QED processes are expected. These manifest in the simultaneous appearance of both the so called above threshold ionization (ATI) [12] and high harmonic generation (HHG) [13,14] processes on atoms or at surfaces. For example, unusually high multiphoton ATI electron yield, furthermore keV electron acceleration phenomena were observed in intense plasmonic fields

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on metal surfaces [15-17] and we predicted recently that with the help of the tunneling emission of electrons in the evanescent field of SPPs the carrier-envelope phase of the few-cycle laser pulses can be controlled [18]. Very recently HHG light was generated on atoms (which is the complementary process of the strong laser field induced electron emission, ATI) using the enhancement effect of the evanescent field [2].

2. Theoretical considerations

In our recent work we used our simple theoretical model to study scattering phenomena in plasmonic fields [4,5]. In a simple way, this model furnishes the main predictions related to the surface plasmon processes for the metal surfaces by introducing the so-called "laser-induced oscillating double layer potential", U_d introduced by us for the first time [4]. We have shown that U_d governs and accounts for all known linear SPP processes, as far as we restrict ourselves to its first approximation term, $U_d^{(1)}$, where we expand U_d according to the displacement of the oscillating electron of the metal. However, to be able to interpret new nonlinear phenomena observed at higher exciting laser fields, we had to generalize the results of Refs. [4,5], by taking the second order term, $U_d^{(2)}$, into consideration in the expansion. This results in $U_d = U_d^{(1)} + U_d^{(2)}$. The detailed description of this generalization will be published elsewhere [6]. Therefore, here we restrict ourselves to the summary of the main predictions of [4,5] and [6] related to the interpretation of our experimental observations. We predicted processes not considered in former models that describe properties of light, emitted by SPPs from thin films [19-21]. The most important statement following from our model, is that the gradient of U_d (along metal surface normal) renders the field enhancement factor in the evanescent region (in the proximity of metal surface) proportional to ω_p^2/ω^2, where ω_p is the plasma frequency of the metal, and ω is the laser frequency. Because $\omega_p/\omega \sim 10$ for the laser field, the field and the intensity enhancement is 100 and 10^4, respectively, in agreement with other theories.

A further consequence is – although only in an implicit form – that the lobe in the angular distribution of this light is sharper than the one found in earlier works [19-21]. It appears at almost tangential emission angles (70°-80°) both around the λ_L wavelength of the exciting laser and at its second harmonic [5,6]. The intensity, the actual emission angle and the width of the lobe of this SPP-emitted light depends not only on the average size of the surface irregularities of the gold film but in addition, on the laser intensity, too. The model also indicates that both perturbative and non-perturbative tunneling processes also occur (depending nonlinearly on the exciting light intensity) in the region of high laser intensities, in addition to the well-known linear responses. Just to name a very recent observation related to this, the occurrence of the nonlinear surface polarization was confirmed by observing SPP excitation by four wave mixing [22].

Therefore, following these considerations and results, in the present work we concentrate on the investigation of linear and nonlinear dependences of the SPP emitted light, generated by the strong evanescent plasmonic field. These are more characteristic, especially for the SPP physics than the usual ATI and HHG effects in previous gas-phase and surface studies.

It should be noted that two other processes may contribute inherently to the observed light at high intensities, in addition to photons emitted by the decaying SPPs. The first one is multiphoton excitation that can lift the d-electrons of the metal into the conduction band. These electrons, when recombining into the hole of the d-band, emit "metallic fluorescent" light, which has a broad spectrum and which is depolarized, in contrast to the SPP-emitted fundamental and second harmonic light. These are expected to be p-polarized similarly to the exciting laser light [23,24]. Secondly, if the laser pulses are extremely short, there is "not enough time" for the electron-phonon interaction in the metal. Therefore, the conduction electron cloud plays a role alone, having its own very low $c_e \sim \gamma T$ specific heat even at the arising high electron temperatures T. This leads to a fast increase of the temperature of this cloud to a high value, which subsequently decreases fast, leading to the emission of

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incoherent, depolarized light with a non-equilibrium Planck spectrum [25,26]. This broad depolarized spectrum can be observed simultaneously with the SPP emitted light and experimental efforts have to be made to separate them.

3. Experimental results

In the experiments presented here, we analyzed the light emitted by SPPs, with the excitation in the Kretschmann geometry. Both angular and spectral distributions of this light were measured on different gold samples, evaporated on glass prisms. A novel, long-cavity, chirped-pulse Ti:sapphire oscillator served as the plasmon exciting light source pumped by a 7-W diode-pumped solid-state laser (Laser Quantum, "finesse"). The oscillator delivered 200 nJ pulse energy with 3.6 MHz repetition rate and the central wavelength of the pulses was λ_L = 795 nm. The outcoupled, highly chirped, 2-3 ps pulses of the laser were extracavity compressed with a pair of transmission gratings to a duration of about 120 fs. A more detailed description of the laser system can be found elwewhere [27,28].

The gold films were evaporated on glass prisms with 45 nm thickness and surface plasmons were excited by p-polarized light. By focusing the laser beam, the intensity could be increased to ~10^{12} W/cm^2 on the surface. Special care was taken to exclude the transmission of all those light contributions which could have hit the Au film at an angle other than the critical plasmon resonance angle. Therefore, before the prism, the incoming laser beam passed through a slit, the edges of which were parallel to the Au film plane. In the totally reflected light spot after the prism the usual dark plasmon absorption line appeared and the width of the slit was adjusted to match the width of this dark line. As a result, at ~0.3 mm slit width only the resonant dark absorption line (the SPP exciting component) was permitted to pass. This layout is illustrated in the inset of Fig. 1(a). The light emitted by the decaying SPPs was detected by a photomultiplier placed at the exit slit of a monochromator. The incident, exciting laser light was p-polarized. The emission angle is measured from the surface normal.

In the first set of our measurements we studied the angular distribution of SPP emitted light as a function of the inducing laser intensity. Two typical examples of the detected angular distribution are shown in Fig. 1(a) (at ~2×10^{10} W/cm² laser intensity, blue dotted curve and at ~5×10^{11} W/cm² laser intensity, red dashed curve). The high intensity distribution can be compared to the equivalent theoretical curve derived from our model for this high-intensity case (Fig. 1(a), black solid curve) [6]. In the lower intensity case the angular distribution of the SPP-emitted light is relatively broad (full width at half maximum (FWHM) is 26°) and exhibits a maximum in the angular range around ~70°. With increasing intensities, however, the maximum is shifted toward larger angles (up to ~82°) and becomes narrower at the same time (FWHM is 12°). The latter values (measured at high intensities) are much lower than those found in earlier works where the measurements were performed at much lower laser intensities [19-21]. By measuring the emission at s-polarization of the incident laser beam, practically no signal could be detected (Fig 1(a), stars). The corresponding scattered spectra from the surface for the high- and low-intensity cases are depicted in Fig. 1(b). There is not any significant difference between the spectral distributions in the two cases.

#101816 - $15.00 USD Received 22 Sep 2008; revised 16 Nov 2008; accepted 24 Nov 2008; published 16 Dec 2008

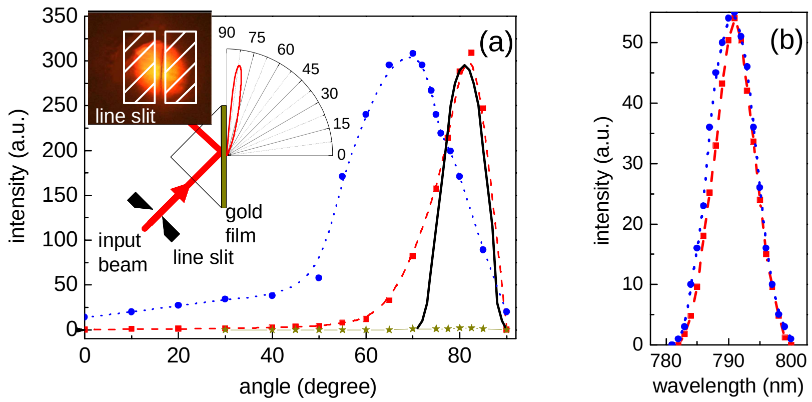

Fig. 1. (a) Scheme of the experimental setup (inset) and angular distribution of the SPP emitted light with 120-fs, p-polarized excitation pulses. The peak intensity was ~2×10^{10} W/cm² (blue circles and dotted curve) and ~5×10^{11} W/cm² (red squares and dashed curve). The distribution for s-polarization (stars) as well as a theoretical curve (thick solid line) are also shown. (b) Spectrum of the SPP emitted light for ~2×10^{10} W/cm² (blue dotted curve) and ~5×10^{11} W/cm² (red dashed curve).

In our second set of experiments, we further increased the I_L intensity of the incoming laser light and assessed the laser intensity dependence of the peak of the SPP-emitted light (Fig. 2). Surprisingly, at around 1.4×10^{11} W/cm^2 peak laser intensity value the curve diverted from the linear dependence, found to be true at lower intensities corresponding to the prediction of [6] suggesting the appearance of the perturbative (nonlinear) behaviour. At these intensity values the U_p ponderomotive potential reaches the $U_p \propto I_L\lambda_L^{\,2}$ ~ 1 eV value if we suppose 1-2 orders of magnitude em field enhancement in the evanescent region. This field enhancement value is in accordance with recent observations [2,3,10,11]. U_p is known to be a decisive parameter for the appearance of so-called strong-field, i.e. non-perturbative interactions and U_p ~1 eV is typically taken to be the strong-field limit.

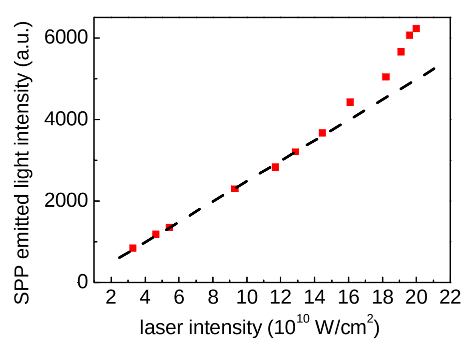

Fig. 2. Intensity dependence of the SPP-emitted light with femtosecond SPP excitation (squares). Nonlinear deviation from the linear fit to the first points (dashed line) starting at 1.4×10^{11} W/cm^2 focused intensity is observed.

In a third set of experiments we also studied the spectral properties of SPP-emitted light around the maximum emission angles. We also observed spectral components at other wavelengths than the fundamental. These results are shown in Fig. 3. Firstly, we observed a

#101816 - $15.00 USD Received 22 Sep 2008; revised 16 Nov 2008; accepted 24 Nov 2008; published 16 Dec 2008

second harmonic peak around $\lambda = 395$ nm which showed a quadratic laser intensity dependence (with an exponent of $n = 2.01 \pm 0.16$), as expected. This light is also p-polarized. The observed half widths of the angular lobes and of the spectral widths are somewhat larger, than those of the fundamental component. The broader angular distribution is in contrast with our prediction. The origin of this phenomenon requires further studies.

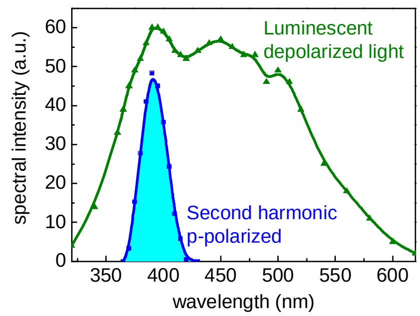

Fig. 3. Second harmonic (blue solid line) and continuum (green solid line) spectra scattered from the metal surface upon SPP excitation with 795-nm, 120-fs pulses.

Fig. 3 also shows a depolarized, very broad spectrum stretching from 300 to 650 nm, (the width of this range is also limited by instrumental parameters: filters, photomultiplier window transmission and sensitivity etc.). This supercontinuum spectrum always appeared as a background in our experiments at the highest laser powers, being 3-4 orders of magnitude weaker than the fundamental component. The intensity integrated for the whole broadband spectrum shows a fourth order laser intensity dependence ($n = 4.06 \pm 0.17$). The angular distribution of this light is isotropic. This behavior can correspond either to the aforementioned ”metallic fluorescence” [23,24] or to the Planck-radiation (following from the Stefan-Boltzmann law [25,26]).

4. Summary

In conclusion, our observations indicate that in SPP-enhanced evanescent electric fields, strong-field optics effects can be observed, which are characteristic for surface plasmon physics, which otherwise occur only in fields, higher than those produced by our exciting laser. These characteristic effects are i) the nonlinear power dependence of SPP emitted light of the exciting laser light above a threshold, ii) the shift of the angular distribution maximum of the SPP emitted light to larger angles with increasing laser intensity, iii) the sharpening of this angular distribution with increasing laser intensity, and finally iv) the occurrence of a broad, isotropic, luminescent or Planck-like spectrum at higher intensities, accompanying the above listed effects. These observations were obtained in a novel way, by combining simple femtosecond oscillators delivering moderate intensities with the strong enhancement effect of the evanescent field of surface plasmon polaritons. SPPs certainly play an important role, serving as novel tools both for fundamental studies in intense field optics and for potential applications.

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