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Experimental and theoretical studies of the statistical properties of surface plasmon polaritons (SPOs) are described. Both classical and non-classical properties of surface plasmons are analysed. The temporal statistical behaviour at low excitation level, as measured by detecting the SPO emitted photon statistics as expressed by the correlation function and the temporal photon count distribution, show that the SPOs preserve the photon statistics of the laser. In the spatial distribution of the plasmon field as measured by an STM, squeezing, i.e. non-classical properties, were found. Independent simple model calculations confirmed the existence of both enhanced EM fields of surface plasmons and their squeezed character.

1. Introduction

Metals have been considered useless in optical applications due to their high absorption. This situation has changed in the last decades due to a 'new type of light', surface plasmon polaritons or briefly surface plasmon oscillations (SPOs).

SPOs are wave-like density fluctuations of conduction electrons on metallic surfaces coupled with a p-polarized electric field in the dielectric material (or vacuum) above the surface. Their properties can be calculated from Maxwell's equations. The condition of their existence is an imaginary metallic dielectric constant with negative real part with an absolute value larger than its imaginary part. The dielectric constant of the dielectric should, of course, be positive.

SPOs have unique properties. They propagate along the metal surface and have dispersion relations which are below the light line in the ω–k plane and are therefore nonradiative. They are not restricted by the diffraction limit and therefore may

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propagate in nanometric structures or be used in sub-wavelength resolution near field microscopy. They represent extremely large (evanescent) electromagnetic fields at the metal surface leading to enhanced surface phenomena especially at so-called hot spots e.g. in Raman scattering. They may have a band gap if excited on an appropriate grating. SPOs may be localized on (metallic) nano-particles and as a special case on the metallic tip of an STM (tunnelling microscope).

These properties, characterizing a 'new type of light' have been broadly studied by many research groups including us, since they offer a broad spectrum of applications in material sciences and nanotechnology [1–5].

In recent years, however, both theoretical and experimental studies indicated that at certain conditions, i.e. in nonlinear cases (as we shall see below in the theoretical part and STM measurements), they may have non-classical properties and could be strong candidates as a 'medium' for quantum information technology applications [6, 7].

According to these arguments, in this paper we report our first theoretical and experimental findings indicating both the classical and nonclassical features in the SPO.

As for our first experiment, we present here briefly the main conclusions only, the details have been described in [8–12]. According to [13, 14], the experiment was performed in the Kretschmann-geometry using gold and silver layers.

The relaxed diffraction limit is the basis of the high-resolution near-field microscopy realized with the help of an STM, where the near field has been realized by SPOs.

This microscope enables us to detect three images simultaneously, namely a topographic, a surface plasmon field and a so-called thermal image which is detected in the absence of the SPO field but reflects its warming up effect due to the decay of the plasmons. A typical example of three such images is shown in figure 1.

The SPO exciting light is generated at 680 nm by a semiconductor laser and the resolution is around 1 nm. The results in figure 1 clearly show the excellent appearance of the SPOs, with the two mentioned necessary controls. These SPOs appear in the strongly enhanced electromagnetic (EM) field above the metal layer.

To interpret this strong EM field enhancement we present below an alternative theoretical model (equivalent with the other existing ones) estimation which, at the same time, also indicates on the example of a nonlinear surface phenomenon (namely multiphoton surface photoeffect, see [15]) the influence of the giant field of surface plasmons. Non-classical properties of SPOs are also expected to follow from this model.

2. Theoretical model calculations to interpret SPO processes

In order to illustrate the appearance of enhanced nonlinearities in the electron excitations due to the enlarged electric field of surface plasmon polaritons, let us first calculate the electron displacements in the bulk of the metal caused by the z component of the penetrating electric field $F(x', z', t) = F_0 \exp(z'/\delta) \sin(\omega t - kx')$.

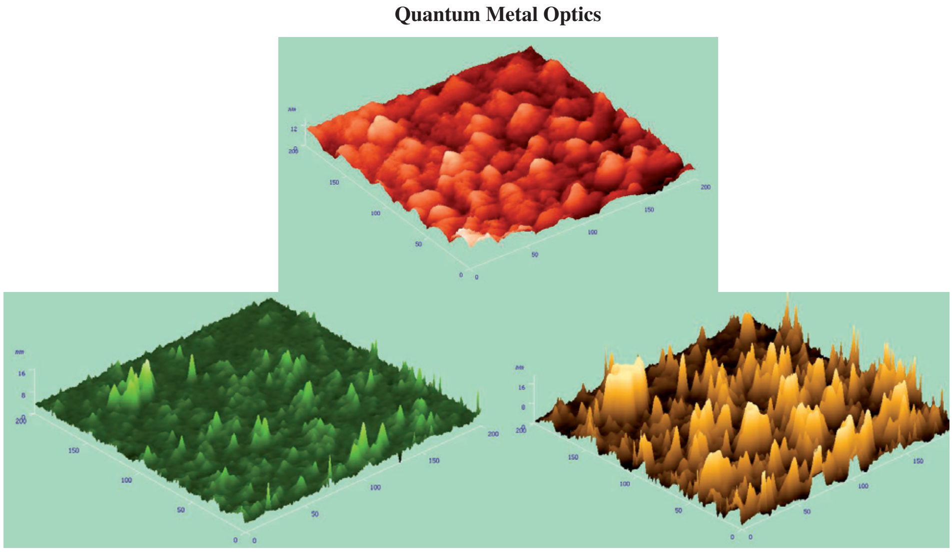

Figure 1. Simultaneously measured topographic, surface plasmon EM field and thermal images obtained by an STM. (The color version of this figure is included in the online version of the journals.)

Here, F_0 and $\omega = 2\pi\nu$ are the peak field strength and the circular frequency of the laser, $\delta = 1/k_m \approx c/\omega_\mathrm{p}$ is the skin depth and $k = (\omega/c)\sin\theta$ denotes the plasmon wave number, respectively. Moreover, we have introduced the plasma frequency $\omega_\mathrm{p} = (4\pi n_\mathrm{e} e^2/m)^{1/2}$, where e and m are the electron's charge and mass, respectively, and n_e denotes the free electron density in the metal. The displacement $\xi(x', z', t) = \alpha_0 \exp(z'/\delta) \sin(\omega t - kx')$ of an electron in the bulk, at an average position (x', y', z'), can be obtained from the solution of the corresponding Newton equation, where the amplitude of oscillation is given as $\alpha_0 = eF_0/m\omega^2$. The potential energy $U_\mathrm{d}(\mathbf{x}, t; \mathbf{x}')$ of a test electron at position $\mathbf{x} = (x, y, z)$ in the joint Coulomb field of a background ionic core at a fixed position $\mathbf{x}' = (x', y', z')$ and of an associated oscillating background electron, is given by $U_\mathrm{d}(\mathbf{x}, t; \mathbf{x}') = e^2/|\mathbf{x} - \mathbf{x}'(t)| - e^2/|\mathbf{x} - \mathbf{x}'|$. Here $\mathbf{x}'(t) = \mathbf{x}' + \varepsilon_z \xi(x', z', t)$ is the instantaneous position of the oscillating background electron, with ε_z being a unit vector perpendicular to the metal–vacuum interface, pointing to the positive z direction. The total potential energy of a test electron is the sum of all the contributions coming from the interactions originating at the positions $\mathbf{x}'$, i.e.

$$U_\mathrm{d}(\mathbf{x}, t) = \sum_{\mathbf{x}'} U_\mathrm{d}(\mathbf{x}, t; \mathbf{x}') \to n_\mathrm{e} e^2 \int \mathrm{d}^3x' \, \frac{\xi(x', z', t)(z - z')}{|\mathbf{x} - \mathbf{x}'|^3} + O(\xi^2). \quad (1)$$

In obtaining equation (1) we have used the continuum limit of the summation and we have expanded the joint Coulomb interaction of the background in powers of the oscillating displacement ξ. The first term on the right-hand side of equation (1) can be calculated analytically,

$$
\begin{aligned}
U_{\mathrm{d}} &= 2\pi n_{\mathrm{e}} e^2 \sin(\omega t - kx) \int_{-\infty}^{0} \mathrm{d}z' e^{z'/\delta} e^{-k|z-z'|} \frac{z-z'}{|z-z'|} \\
&= U_{\mathrm{D}} \sin(\omega t - kx) \begin{cases} \dfrac{2e^{z/\delta}}{1-k^2\delta^2} - \dfrac{e^{kz}}{1-k\delta}, & (z<0), \\ \dfrac{e^{-kz}}{1+k\delta}, & (z>0). \end{cases}
\end{aligned} \tag{2}
$$

$$
\begin{aligned}
U_{\mathrm{D}} &\equiv (\omega_{\mathrm{p}}/2\omega)^2 (\delta/\lambda)\mu(2mc^2), \\
\mu &\equiv eF_0/mc\omega = 10^{-9} I^{1/2}/E_{\mathrm{ph}}, \\
\mu^2 &= 10^{-18} I\lambda^2.
\end{aligned} \tag{3}
$$

In equation (3) we have introduced the amplitude U_{D} of the oscillating collective potential energy of the test electron, equation (2), which can take on very large values even for relatively moderate laser intensities (notice the factor $2mc^2$ being just the pair-creation energy $\approx 10^6$ eV). The dimensionless intensity parameter μ usually shows up in any strong field calculation, its magnitude governs the nonlinearity of direct laser–electron interactions. In equation (3) I denotes the peak laser intensity in $\mathrm{W\,cm^{-2}}$, E_{ph} is the photon energy measured in eV and λ is the central wavelength in microns (10^{-4} cm). We note that the gradient of the potential energy, equation (2), essentially equals the force acting on a test electron due to a corresponding surface plasmon polariton electric field. For $|kz|$, $|kx| \ll 1$ and $|z/\delta| \gg 1$ the potential energy in equation (2) can be well approximated in the vicinity of the metal surface by the following double-layer potential energy $U_{\mathrm{d}} \approx \mathrm{sign}(z) U_{\mathrm{D}} \sin(\omega t)$, where $\mathrm{sign}(z>0) = 1$ and $\mathrm{sign}(z<0) = -1$, thus the maximum total jump in the energy equals $2U_{\mathrm{D}}$. Because of this property, henceforth we will call U_{d} '*the laser-induced oscillating double-layer potential*'.

The concept of the laser-induced oscillating double-layer potential outlined above was first introduced in our earlier study [1] in order to explain a surprising outcome of one of our experiments [15], namely, the appearance of very large ($\sim$600 eV) energy photoelectrons induced by Nd:Glass laser radiation ($h\nu \sim 1.17$ eV) at moderate intensities of some $10\,\mathrm{GW\,cm^{-2}}$. There the main problem was that the very large nominal order of the photon absorption processes corresponding to the experimental results ($n \sim 5$–600) could not have been deduced even from the usual non-perturbative approach based on Gordon–Volkov states [16], since the intensity parameter $\mu = eF_0/mc\omega$ was very small, of order of 10^{-4} in that case. That time we realized that instead of μ another basic dimensionless parameter 'a' appears in the analysis in a natural way, when we introduce the interaction with the double-layer potential, which builds up due to the coherent collective excitation of all the electrons within the skin depth. The parameter a is defined as $a = 2U_{\mathrm{D}}/h\nu$, where $U_{\mathrm{D}} \approx (\omega_{\mathrm{p}}/2\omega)\mu mc^2$ is the amplitude of the double layer potential energy of a test electron. The size of this a governing the degree of nonlinearity turned out to be just of order of 500 for the mentioned experiment, hence we were able to explain the basic features of the measured electron spectra.

In the present section we apply the above-mentioned model for a different situation in order to see whether we can interpret another strange experimental result [17] concerning electron emission from gold cathodes (work function $\sim$5 eV) irradiated by mid-infrared radiation (generated by the Orsay Free Electron Laser) of wavelength up to 12 μm ($h\nu \sim 0.1$ eV) in the $I \sim 10\,\mathrm{MW\,cm^{-2}}$ intensity regime. The intensity parameter is extremely small in this case: $\mu \approx 3 \times 10^{-5}$. The minimum number of photons required for the deliberation of an electron from the binding is of order of 50. As was pointed out by the authors of this paper, both the tunnelling model and the multiphoton model predict numbers many (about 200) orders off the experimental figures. For an explanation they have introduced the concept of 'lucky electrons', based on a classical acceleration mechanism. We give an alternative quantum-mechanical description based on our double-layer model (see figure 2). Since in the present case the mean collision frequency of the electrons is comparable with the frequency of the radiation, the electrons in the bulk do not contribute to the collective coherent excitation of the double-layer potential. Only evanescent electrons in a thin layer in the immediate vicinity of the surface make a contribution. Under this circumstance, the amplitude of the potential energy is of order of

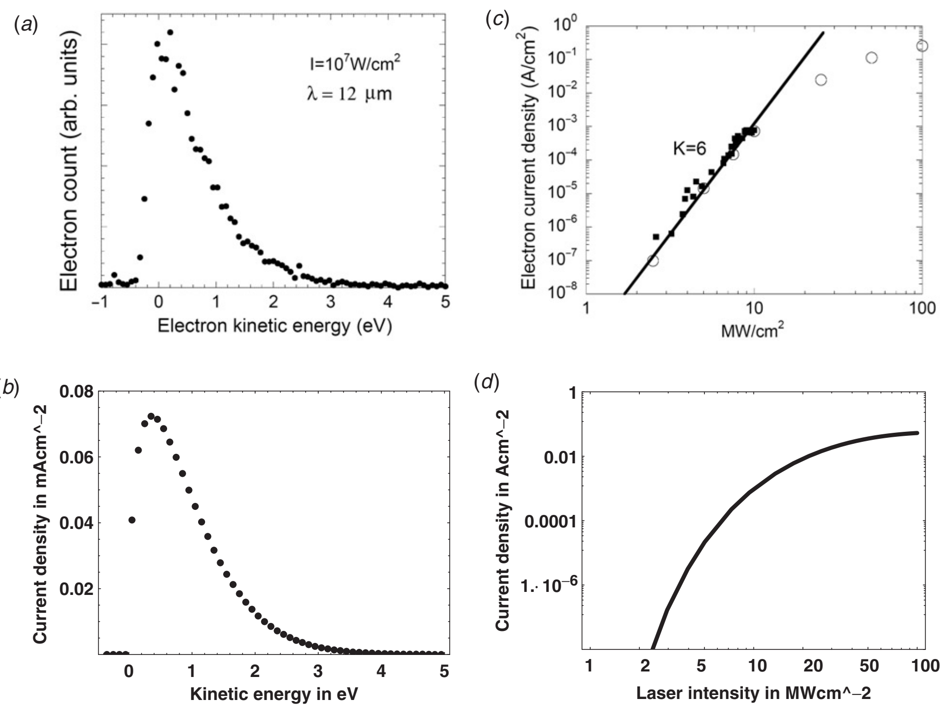

Figure 2. Experimental (*a*) and theoretical (*b*) photoelectron spectra at 100 MW/cm^2 laser intensity and 12 microns wavelength and the intensity dependence of the total photocurrent density according to the experiment (*c*) and the present theory (*d*).

$U_D \approx 1.4\,\mathrm{eV}$. By using the Floquet method we have found an approximate analytic solution of the quantum-mechanical barrier problem of the electrons scattered by the oscillating double-layer potential at the metal surface. We have checked that the exact multiphoton transmission coefficients calculated numerically can be well approximated by ordinary Bessel functions, i.e. $|T_n|^2 \approx J_n^2(a)$. By averaging over the amplitudes of the laser pulses with respect to the Gaussian distribution $\exp(-F^2/F_0^2)/F_0^2$, we have found that the multiphoton current components are governed by the formula $j_n \sim p_n I_n(a_0^2/2)\exp(-a_0^2/2)$, where I_n denotes the nth order modified Bessel function of the first kind, and the nonlinearity parameter equals $a_0 = 2U_D/h\nu = 28$ in the present case. The momentum $p_n = (-A + nh\nu)^{1/2}$ corresponds to n-photon absorption, where $A \approx 5\,\mathrm{eV}$ is the work function of the gold target. The theoretical results based on this formula reflect excellently back all the characteristics of the observed photoelectrons, namely the unexpectedly wide above-threshold spectrum extending up to 2 eV (corresponding to $\sim 50 + 20 = 70$ photon absorption), the intensity dependence d log j/d log $I \sim 6$ and the very high magnitude of the total photocurrent $j \sim 1\,\mathrm{mA\,cm^{-2}}$.

At the end of the present section let us note that if in equation (1) we take the quadratic term also into account, then, in the quantized description of the photon field, the interaction Hamiltonian will contain a term proportional to $(\hat{a} + \hat{a}^+)2$, with $\hat{a}$ and $\hat{a}^+$ being the photon annihilation and creation operators, respectively. In the present section we have discussed the problem of very high-order nonlinear *electron signals* induced by a *classical electromagnetic radiation field* at the metal surface. A preliminary version of this analysis has already been presented in 2000 by one of the authors [18] at an international conference. Later, in 2002, Georges [19] published a thorough quantum mechanical analysis of the same experiment we discussed above. His theory is based on the density matrix formalism which, after all, leads to the rate equations of the step-like excitations of a single electron. According to his results the high-energy above-threshold electrons appear due the step-like absorptions in the continuum due to the presence of the image potential outside of the metal. Our method basically differs from his, because we first calculate the collective response (oscillating double-layer potential) of the bulk electrons classically, and then use quantum mechanics to describe the scattering of one test electron (which can, of course, be any of the electrons).

Concerning the possibility of the appearance of *non-classical light signals* stemming from the metal target (during the plasmon decay), let us note the following. If we take into account the quantized nature of the exciting field, then, roughly speaking, the oscillating part of the 'trajectories' of the bulk electrons (denoted by ξ at the beginning of the present section), is obtained from the Heisenberg equation instead of the Newton equation. Thus, instead of $\sin(\omega t - kx')$ we have $\mathrm{i}[a\exp(-\mathrm{i}\omega t + \mathrm{i}kx') - a^+\exp(+\mathrm{i}\omega t - \mathrm{i}kx')]/2$, with a and a^+ being the annihilation and creation operators of the incoming quantized mode, respectively. Consequently, if in equation (1) we take the quadratic term also into account, then, the collective potential will contain a term proportional to $[a\exp(-\mathrm{i}\omega t + \mathrm{i}kx') - a^+\exp(+\mathrm{i}\omega t - \mathrm{i}kx')]^2$. In this higher approximation squeezed light can be expected from the radiative decay. As we shall see in the next section, for the low intensities we used in our experiment, the size of such a non-classical effect is definitely smaller than 2%. We would like to emphasize that this is a very sketchy argumentation, with the help of which we merely wanted to illustrate that a *bilinear interaction term* can be derived in the frame of our model. And, as we know, on the basis of bilinear interactions several non-classical effects can be produced (see e.g. the excellent collection of papers in the book edited by Dodonov and Man'ko [20]). We note that the nonperturbative description of the interaction of a single electron and a quantized mode of the radiation field has been first analysed by one of the authors [21] and later by Becker *et al.* [22]. The production of squeezed states in the interaction between electromagnetic radiation and an electron gas has been considered by Ben-Aryeh and Mann [23]. Concerning other non-classical effects in the context of plasmon interactions see e.g. the recent studies by Altewischer *et al.* [24].

3. Experiments on statistical properties of SPO

Here we describe our investigations indicating the non-classical properties of SPOs [13]. Two sets of experiments were carried out. In the first one a large number of simultaneous SPO and thermal images like those shown in figure 1 were compared. Statistical analysis of the signal amplitudes in 256×256 pixels of the two types of images were performed and a typical result is shown in figure 3.

The thermal image after the laser pulse, i.e. without SPO, as expected, has Boltzmann-distribution with a cut-off at low amplitudes. This is due to the contact potential between the metallic tip and the metal surface. The distribution of signal amplitudes in the SPO case is Gaussian (the high number limit of a Poisson distribution) but with a sub-Poissonian width indicating a squeezing effect. Although the exciting light intensity used was low, the indication of this appearance of a squeezing effect is attributed by us to the strong nonlinearity, inherent with the STM detection procedure.

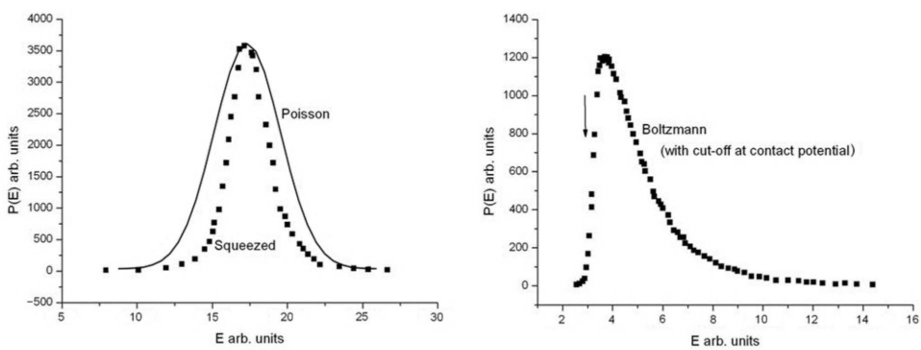

Figure 3. Statistical analysis of the signal amplitudes of the 256×256 pixels of the two type of images (with SPO and without).

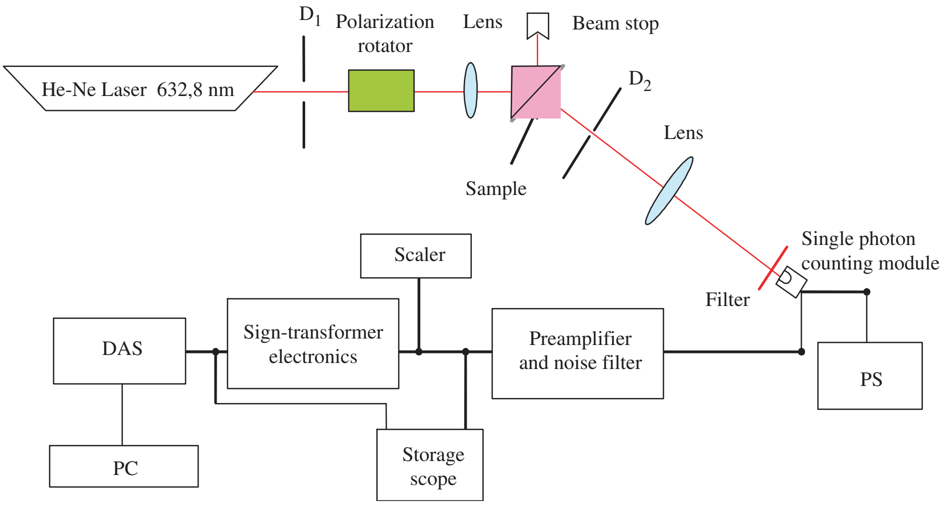

Figure 4. Experimental set-up for measurement of photon statistics of surface plasmons. (The colour version of this figure is included in the online version of the journal.)

The images in this measurement represent temporal averages. Therefore, we decided to measure the time statistics too by comparing the statistics of the exciting laser photons with those emitted by the decaying SPOs.

In this second set of experiments the statistical properties of the light generated by plasmons again at low exciting laser intensity were measured by the following experimental set-up where nonlinear properties are not expected. The optical part of the set-up (figure 4) is nearly the same as in our previous experiment [14]. The detector was replaced by a Perkin Elmer Peltier cooled, temperature stabilized fast SPCM AQR 14-13210 single photon counting module having high quantum efficiency and very low dark count rate. The electronic and data evaluation was replaced by a PC controlled data acquisition unit consisting of noise filtering, signal transforming parts, a National Instruments type DAQ 6602 system, scalers and a controlling PC.

The benefit of the present equipment is the extremely high signal/noise ratio measured at different (S and P) polarization of the incident laser light when plasmons are and not generated (effect, no effect). Due to the precise alignment and optical filtering this ratio (determining the errors of the measurement) at the detector was $>70:1$. The measurement was performed in a pre-programmed regime using a PC as a controller.

The results of the measurement of most relevant statistical functions: correlation function $-G(\tau)$, time interval distribution between the neighbouring photons and number distribution of the detected photons in definite (preset) sampling time, for the exciting laser beam (laser) a light emitted by generated plasmons (effect) are shown on figure 5.

The negligible deviation from 0 level below 2 μs corresponds to afterpulsing of the detector which depends on the breakdown voltage. As predicted in the previous

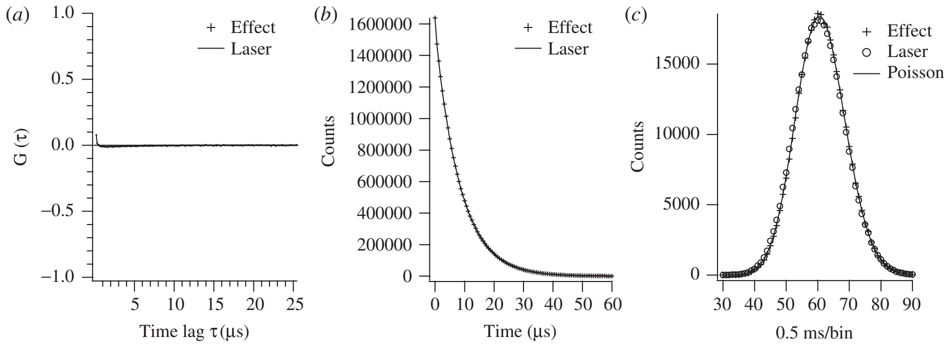

Figure 5. Correlation function (*a*), time interval distribution between the neighbouring photons (*b*) and the number distribution of the detected photons in definite (preset) sampling time (*c*) of the exciting laser beam (laser) and the light emitted by generated plasmons (effect).

theoretical section, the experimentally detected nonclassical effect, if any, must be smaller than 2%.

4. Conclusions

In our measurements at low exciting laser intensities, where we detected the SPO emitted photons, the surface plasmons preserved the temporal statistics of the exciting laser light. In contrast, in the measurements where the electric field of SPOs was detected by an STM, the spatial distribution of this field indicates some non-classical properties of these SPOs. These observations are in line with our model calculations. Future experiments are planned to study the temporal statistics at high excitation levels, where the non-classical properties are expected to show up in the temporal statistics too.

Acknowledgements

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References

[1] S. Varró and F. Ehlotzky, Phys. Rev. A **57** 663 (1998).
[2] N. Kroo, Zs. Szentirmay and J. Félszerfalvi, Phys. Lett. **81**A 399 (1981).
[3] N. Kroo, Zs. Szentirmay and J. Félszerfalvi, Phys. Lett. **101**A 235 (1984).
[4] N. Kroo, Zs. Szentirmay and J. Félszerfalvi, Phys. Lett. **88**A 90 (1982).
[5] H. Raether, *Surface Plasmons* (Springer, Berlin, 1988).
[6] N. Kroo, J.P. Thost, M. Völker, *et al.*, Europhys. Lett. **15** 289 (1991).

[7] S. Fasel, M. Halder, N. Gisin, *et al*., New. J. Phys. **8** 13 (2006).
[8] A. Czitrovszky, A. Sergienko, P. Jani, *et al*., Laser Phys. **10** 86 (2000); A. Czitrovszky, A. Sergienko, P. Jani, *et al*., SPIE **3749** 422 (1999).
[9] D. Oszetzky, A. Czitrovszky and A. Sergienko, SPIE **5161** 352 (2003).
[10] D. Oszetzky, P. Gäl and A. Czitrovszky, J. Aerosol Sci. **33** 278 (2004).
[11] D. Oszetzky, Quantum Electron. APH B **20** 185 (2004); D. Oszetzky, A. Czitrovszky and A. Nagy, SPIE **6028** 449 (2006).
[12] D. Helitmann, N. Kroo, Zs. Szentirmay, *et al*., Phys. Rev. B **35** 2660 (1987).
[13] N. Kroo, Zs. Szentirmay and H. Walther, Surf. Sci. **582** 110 (2005).
[14] N. Kroo, A. Czitrovszky, A. Nagy, *et al*., J. Mod. Opt. **53** 2309 (2006).
[15] Gy. Farkas and Cs. Tóth, Phys. Rev. A **41** 4123 (1990).
[16] N.J. Kylstra, C.J. Joachain and M. Dörr, in *Atoms, Solids and Plasmas in Super-intense Laser Fields*, edited by D. Batani, C.J. Joachain, S. Martellucci and A.N. Chester (Kluwer Academic/Plenum Publishers, New York, 2001), pp. 15–36.
[17] Gy. Farkas, Cs. Tóth, A. Kohäzi-Kis, *et al*., J. Phys. B: At. Mol. Opt. Phys. B **31** L461 (1998).
[18] S. Varró, in Book of Abstracts of the 9th Annual International Laser Physics Workshop (LPHYS 2000), Bordeaux, France, 17–21 July (2000).
[19] A.T. Georges, Phys. Rev. A **66** 063412 (2002).
[20] V.V. Dodonov and V.I. Man'ko (Editors), *Theory of Nonclassical States of Light* (Taylor & Francis, London, New York, 2003).
[21] J. Bergou and S. Varró, J. Phys. A: Math. Gen. **14** l469 (1981).
[22] W. Becker, K. Wódkiewicz and S. Zubairy, Phys. Rev. A **36** 2167 (1987).
[23] Y. Ben-Aryeh and A. Mann, Phys. Rev. Lett. **54** 1020 (1985).
[24] E. Altewischer, M.P. van Exter and J.P. Woerdmann, Nature **418** 304 (2002).