Can the ghost imaging increase the lateral resolution of surface plasmon resonance microscopy?

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Abstract. Surface plasmon resonance (SPR) microscopy is one of the most sensitive optical label-free methods of microscopy. Nevertheless, it does not have a sufficiently high lateral resolution in comparison with other methods of optical microscopy. By analogy with the scattering medium, surface plasmon polaritons (SPPs) blur the observed area. To eliminate this disadvantage, we propose to adapt the method known as ghost imaging (GI), which is notable for its tolerance to environmental aberrations between an object and a camera. In this article, we propose a ghost imaging scheme for a surface plasmon resonance microscope using a pseudo-thermal radiation source. We make a fundamental analysis of the factors affecting the resolution capability of the ghost SPR microscopy. We claim that applying the ghost imaging method to SPR microscopy can improve its lateral resolution by eliminating uncorrelated with modulated radiation phase noise generated by the process of random re-emission of surface electromagnetic wave (SEW) from the site of excitation. In combination of factors, the ghost imaging method of SPR microscopy is potentially capable of becoming outstanding among other methods of microscopy of thin films.

1. Introduction
SPR microscopy is a label-free optical method of microscopy of thin dielectric films with phenomenal sensitivity among other reflection methods (such as differential interference contrast microscopy [1], backside absorbing layer microscopy [2] and others [3]) and ultrahigh vertical resolution reaching $\lambda / 100$. Due to the simplicity of the method, SPR microscopy is widely used in non-destructive testing of thin films, biology, and medicine [4], [5]. However, as was noted more than once by the researchers [6], starting with the author of the method [7], [8], the disadvantage of SPR microscopy is low lateral resolution, which prevents it from becoming outstanding among existing methods of microscopy of thin films (such as atomic force microscopy and other scanning methods).

2. Principles of SPR microscopy
SPPs are not real particles, but quasiparticles (quanta of collective vibrations) arising as a result of oscillations of surface charges (conduction electrons in the skin layer of the metal) relative to the ionic core of the metal crystal lattice (or semiconductor). Their existence follows from nonradiating solutions of wave equations in the form of surface electromagnetic waves at the interface between media with different signs of the real part of the complex permittivity. The SPPs localization region looks like evanescent waves. Their energy increases exponentially near the surface. Therefore, the
interaction of light with the surface increases significantly and determines the sensitivity of the SEW to any slightest change in the dielectric constant or surface topography.

Note that SPPs have a boundary frequency. This is due to the fact that surface excitations exist only as long as the generation of volumetric excitations is difficult. The boundary frequency between the excitation of surface and volume waves for plasmons is the fundamental frequency of the electron gas of the metal (plasma frequency $\omega_p/\sqrt{2}$). At frequencies higher than boundary $\omega_p/\sqrt{2}$ surface excitations (SPP) are not possible. Moreover, the wave vector of the SPP propagating along the surface of the metal differs from the wave vector of the volume plasmon. The relationship between the SPP wave vector $k_x$ and the SPP frequency $\omega$ (quantum energy) is described by the dispersion relation [9]:

$$k_x = \frac{\omega}{c} \left( \frac{\varepsilon_d}{\varepsilon_d + \varepsilon_m} \right)^{1/2},$$

where $c$ is the speed of light, $\varepsilon_d$ and $\varepsilon_m$ are the complex permittivities of the dielectric and metal layers.

It follows from the formula that each frequency of light corresponds to its own wave vector for the excitation of the SPP. This method of excitation is called resonant, since in the case of illumination of the surface by a non-monochromatic light beam, only light quanta, for which the laws of conservation of energy and momentum are satisfied, will excite the SPP. To fulfill the conditions for matching wave vectors, there are various light input schemes such as lattice, end-fire coupling and prism, called attenuated total reflection (ATR) methods. The most popular ATR scheme is the Kretschmann scheme. In the case of ATR, plasmon resonance microscopy is based on determining the reflection coefficient of light incident at an angle greater than the angle of total internal reflection in the prism. The reflection coefficient decreases when the excited SPPs carry away part of the energy of the incident light. The dependence of the reflection coefficient on the angle of incidence of light is shown in Fig. 1 and is described by the equation:

$$R(\theta) = 1 - \frac{a^2}{(\beta - \beta_r)^2 + a^2},$$

where $\theta_r$ is the resonance angle, $\alpha$ is the excitation coefficient of the SPP, $\alpha$ is the half-width of the resonance curve [7].

Figure 1. An example of a resonance curve.

Figure 1 shows that the excitation of the SPP occurs not at a certain angle of incidence, but at a set of angles. This set of angles corresponds to a set of photon pulses (a set of wave vectors). The reason for this is the finite lifetime of the SPP. The large width of the resonance curve corresponds to a large spread in the SPP energy or their short lifetime. The longer the lifetime of a plasmon, the stronger the interaction is. Therefore, a greater amount of energy is converted to a SPP and thus, the vertical resolution becomes higher.
Many factors influence the lateral resolution of SPR microscopy. The most important physical reason is that the area of localization of the SPPs on the surface exceeds the area illuminated by the exciting light beam, which introduces spurious illumination. SPPs propagate from the excitation region along the interface and are randomly re-radiated (see Fig. 2). This additional illumination “blurs” the resulting image of the surface. The distance over which they propagate is determined by losses due to scattering on the roughness of the metal film and absorption. The average distance at which the SEW intensity decreases by a factor of $e$ (the propagation length $L_x$ of SPP) is defined as [9]:

$$L_x = \frac{1}{2k'_{x}},$$

where $k'_{x}$ is the imaginary part of the SEW of wave vector. Moreover, $L_x$ can reach macroscopic distances (much larger $\lambda$). This is similar to the interaction of SPP on metal for THz [10].

![Figure 2. Schematic diagram of the mechanism of SPP reemission outside the excitation point.](image)

- $I_i$ – incident radiation, $I_r$ – reflected radiation, $I_s$ – scattered radiation from the SPP.

Obviously, the higher the frequency, the smaller the distance SPPs can overcome and they are localized when approaching the resonant frequency. However, near the resonant frequency, the sensitivity of the method decreases. Therefore, traditional SPR microscopy is forced to seek a compromise between vertical and lateral resolution by selecting the optimal substrate material, its optimal thickness, and the size of the scanning beam. Since the resonance frequency of most metals lies in the UV, the SEWs at THz frequencies are very far from resonance and have macroscopic distances (hundreds of wavelengths) of propagation length. Therefore, to reduce the propagation length in THz range, one can use semiconductors with various doping, for example, InSb [11].

Thus, a surface electromagnetic wave (SEW) excited by an incident beam can accidentally re-radiate from the surface around the point of incidence of the beam and erodes the observed region in which a response to SEW excitation is expected. One can compare the collective oscillations of the SPP with the role of the scattering medium between the object and the camera. The maximum achievable lateral resolution is limited by the propagation length of the SEW. There is a method resistant to aberrations between the object and the camera, such as turbulent and scattering media, known as ghost imaging (GI) [12]. This method is potentially capable of removing the effect of illumination from reradiated SPPs in SPR microscopy.

3. Adaptation of ghost imaging to SPR microscopy

The GI method established itself in the classical optical visible-range nanoscopy [13], allowing to obtain a high resolution. In GI the image of an object $O(x,y)$ is reconstructed by calculating the second-order correlation function (mutual intensity) between the spatial distribution of the intensity
\( P(x, y) \) of the probing beam (masks) and the integral intensity \( S \) of the reflected (or transmitted) light registered by a single-pixel receiver \([14]\) in two optical arms \((x, y)\) and \((x', y')\):

\[
O(x, y) \propto \sum_{i=1}^{N} (S_i - \langle S \rangle)(P(x, y)_i - \langle P(x, y) \rangle) = \langle P(x, y)_i \cdot S_i \rangle - \langle P(x, y) \rangle \langle S \rangle , \quad (4)
\]

where \( S = \int R(x', y') P(x', y') \, dx' \, dy' \), \( \langle \ldots \rangle = \frac{1}{N} \sum_{i=1}^{N} \ldots \) is the averaging operator, \( N \) is the number of independent patterns \( P(x, y) = P(x', y') \), \( R(x', y') \) is the object response function.

The restored image is called a ghost image, because it uses information about the spatial structure of light that did not interact with the object. According to the method of creating a correlation between the optical arms, the GI method is divided into quantum, classical, and algorithmic. Initially, GI was implemented in the quantum case, and then demonstrated using classical (thermal and pseudo-thermal) sources, and then algorithmic devices like spatial light modulators (SLM) \([15]\).

Taking into account our practical interests in THz optics, we note that GI has great potential in the electromagnetic spectrum regions, where detectors are quite complex and expensive, such as in the terahertz (THz) or far infrared (IR) range \([16]\). At the moment, neither the quantum nor the classical method of THz GI in the far wave zone has been demonstrated yet. To date, only an algorithmic method of the GI method using pre-made random stencil masks has been demonstrated in the THz range. Also imaging systems using structured metamaterials have been demonstrated \([17]\). However, all THz GI experiments were not able to achieve acceptable image quality, due to the lack of high-resolution spatial modulators for the THz range and the use of low-power THz radiation sources.

Due to the imperfection of algorithmic devices for GI in the THz range and the low efficiency of the nonlinear SPDC crystals \([18]\) for the quantum method, we will consider the classical GI. The classic GI uses a pseudo-thermal light source. The spatial coherence of this source is comparable to a heat source, but the temporal coherence significantly exceeds it. Such a source is, for example, the light of a coherent radiation source transmitted through an inhomogeneous medium. As a result of this, a pattern of random interference – speckle structure – is observed. The classical GI has not been implemented in the THz range yet, since it requires a powerful source of terahertz radiation. To implement this method, we suggest using the Novosibirsk free electron laser as a radiation source, which is characterized by a high beam power (up to 100 W) and a large coherence length (3 cm) of the emitted radiation \([19]\). The high power of the beam provides a speckle structure with a pronounced profile, which is necessary to achieve a high level of contrast in the resulting image. A large coherence length is necessary to ensure a high degree of correlation between the emitted radiations coming out of the beam splitter, which allows you to restore the image with less noise. The THz radiation intensity is modulated using speckle structures used as random masks.

We propose to enhance the classical scheme of the SPR microscopy with a second optical arm (Fig. 3), as in the case of the optical GI microscope \([20]\). To observe plasmon resonance, we select the Kretschmann scheme. To excite the SPPs, we use a pseudo-thermal radiation source. It is created using a laser whose radiation is scattered on an inhomogeneous mirror surface. The S-polarized radiation is cut off to observe resonance on the prism by the ATR method and is collected using the L1 lens. Then the radiation is recorded using a single-pixel detector. We compare the signal from the single-pixel receiver with information about the surface illumination from the prism in the second optical arm (the conditions for total internal reflection without loss are fulfilled on the second prism). To obtain an image corresponding to the illumination of the surface of the second prism, we use the 4f-scheme of the telescope (lenses L2 and L3) and then, rotate the camera so that the image plane coincides with the object plane.
Figure 3. Optical design of a two-arm SPR microscope using GI:

*p-Pol* is polarizer transmitting p-polarized radiation (relative to the surface of the prisms) from the source (for example, THz radiation of *FEL* (Free Electron Laser); *D* is aperture; *BS* is non-polarizing beam splitter; *Prism* are isosceles prisms, a thin film (object) is applied to the base of one to excite the SPP; *Det* is a single-pixel detector (e.g., a Golay cell); *Cam* is a multi-pixel camera (e.g., a matrix of microbolometers); *Li* are collecting lens; *P* is a pinhole diaphragm for cutting off out-of-focus light.

In the case of SPR microscopy, the signal of a single-pixel detector S is written as:

\[
S_l = \int R(x',y')(P(x',y')_l + I_s(x',y')_l)dxdy' = S_o + S_c_l, \tag{5}
\]

where \(P(x,y)\) is light intensity distribution on the surface of a thin film (or an object under study), \(R(x',y') \leftrightarrow R(\theta,\varphi) = \int_{\theta_1}^{\theta_2} R(\theta) \cdot I_l(\theta) + \int_{\varphi_1}^{\varphi_2} R(\varphi) \cdot I_l(\varphi)\) is reflection coefficient obtained by convolution of the resonance curve and a beam of equal intensity \(I_l(\varphi) = I_l(\theta)^2 + I_l(\varphi)^2\) with a finite angular width of the beam at spatial angles \((\theta_1,\theta_2), (\varphi_1,\varphi_2)\). \(S_o\) is the object signal, \(S_c\) is the scattered radiation signal. Obviously, the spurious illumination from SPPs excited at point \((x_1',y_1')\) and randomly reradiated at a point on the surface \((x_2',y_2')\) does not depend on the intensity of radiation incident on point \((x_2',y_2')\) modulated by random speckles:

\[
\langle P(x,y) \rangle_l \cdot I_s(x',y')_l = \langle P(x,y)(I_s(x',y') \rangle. \tag{6}
\]

Then substituting (6) in (4) and using (5) the image can be found as:

\[
O(x,y) \propto \langle P(x,y) \rangle_l \cdot (S_o + S_c_l) - \langle P(x,y) \rangle_l (S_o + S_c_l) = \\
\langle P(x,y) \rangle_l (S_o) + \langle P(x,y) \rangle_l (S_c) - \langle P(x,y) \rangle (S_o) + \langle P(x,y) \rangle (S_c) = \\
O_o(x,y) - \int R(x',y')(P(x,y)_l \cdot I_s(x',y')_l)dxdy' + \langle P(x,y) \rangle (S_c) = \ O_o(x,y). \tag{7}
\]

Thus, we show that the GI eliminates the effect of spurious illumination from re-radiation of SPPs.

4. Discussion

If we do not consider the noise component, the image of the surface \(O_o(x,y)\) is a convolution of the reflectivity of the surface \(R(x',y')\) with the autocorrelation function \(C_{ss}(x-x',y-y')\) of the stationary pattern of the intensity distribution \(P(x,y)\) [21]. In the cases of computational and quantum GI \(C_{ss}(x-x',y-y') = \delta(x-x',y-y')\). This means that in these cases, the lateral resolution of the ghost SPR microscopy is diffraction-limited by the optics of the microscope.
In the case of the classical GI, the surface of the prism is illuminated by an inhomogeneous random interference patterns — speckles. In this case, \( G(x-x', y-y') \) is a Gaussian function \([22]\), which introduces a gaussian blur into the final image. Thus, according to the Nyquist theorem, we cannot distinguish the details of an object less than some "cutoff" spatial frequency that is inversely proportional to the speckle size \( b_s \) (in the case of objective speckles \( b_s \approx 1.5 \frac{\lambda D}{\mathcal{D}} \)). In this case, the minimum speckle size is limited, because we cannot significantly increase the diameter of the aperture \( D \), since this increases the angular divergence of the illuminating beam. Based on the Nyquist theorem, we restrict the angular divergence of the illuminating beam to the half-width \( \Delta \theta \) of the resonance curve \( R(\theta) \). This condition for the angular aperture of the beam \( \alpha = \Delta \theta \) is the main limitation on the lateral resolution of the image, which can be restored using speckles (quasi-thermal light). This limitation corresponds to the flat section of the resonance curve \( R(\theta) \) near the resonance angle \( \theta_r \), when radiation frequencies close to the surface plasmon resonance frequency, at which the reflection coefficient \( R = b_{\text{mean}} = \text{const} \) can be considered approximately constant. But, as in the case of traditional SPR microscopy, this condition leads to a decrease in image contrast \([6]\).

5. Conclusion
It was shown that GI method can increase lateral resolution in SPR microscopy. Therefore, the GI method was adapted for the optical scheme of the SPR microscope, supplementing it with a second optical arm. It is shown that the GI method is capable of eliminating spurious illumination from reradiated SPPs by cutting off random components of reflected radiation uncorrelated with modulated light.

We note that the GI method allows to increase the measurement rate, in contrast with scanning systems \([23]\). It is also possible to improve lateral resolution by increasing the numerical aperture \((NA = n \sin(\alpha))\) in the object arm compared to optical microscopy. Lateral resolution also can be enhanced using materials with a higher refractive index \( n \).

In addition, from the analysis of the detectability of THz detectors \([24]\), it can be concluded that single-pixel receivers (for example, an optical acoustic Golay cell or detectors using hot electrons) have greater sensitivity than multi-pixel receivers (for example, microbolometer arrays). Ghost imaging allows the use of more sensitive single-pixel receivers without resorting to mechanical scanning. As a result, the proposed method of SPR microscopy using GI is potentially able to become outstanding among other methods of microscopy of thin films.

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