LETTER

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Plasmonic dynamic screening in a gold film by intense femtosecond laser light

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Abstract – It has been found experimentally that in intense femtosecond laser fields the surface plasmon dispersion has an oscillatory character as a function of the exciting laser intensity. It has been interpreted as the result of the dynamic screening of electrons by the strong laser field. A simple model is described in addition to the experimental results, being in good agreement with these findings. The results imply an electron effective mass of around 10 percent lighter than the free electron mass. The effective mass decreases with increasing laser intensity.

Plasmonic phenomena in general and nonlinear plasmonics in particular are increasingly in the center of research interest [1–8]. This tendency has been motivated by the several unique properties of bulk but even more by surface plasmons (SPPs) and by potential applications [9–12].

In this spirit we have performed experiments, studying the response of a scanning tunneling microscope (STM) to SPP near fields in a room temperature gold film. SPPs have been excited by a Ti:Sa femtosecond lasers ($\lambda = 805$ nm, pulse length 40–100 fs) [13] via a glass prism in the Kretschmann geometry. In addition multiphoton (multi SPP) electron emission experiments have also been performed, looking for the spectrum of emitted electrons by the time-of-flight method. In both cases the laser intensity has been changed in the ∼20–200 GW/cm^2 range. In a broad intensity range around 80 GW/cm^2 anomalies have been found, interpreted as the consequence of electron pairing, Meissner effect and anomalous Faraday rotation of the polarization plane of light [14–16].

In our measurements, however, all the observed laser-intensity–dependent effects have been modulated periodically as a function of this laser intensity, with a periodicity of about $1.7 \cdot 10^6$ V/cm laser field strength.

The set-up used in our two sets of experiments is shown in fig. 1. Here the angular shift of the plasmon resonance angle has been measured as a function of the laser intensity. The average of several of our measurements is shown in fig. 2.

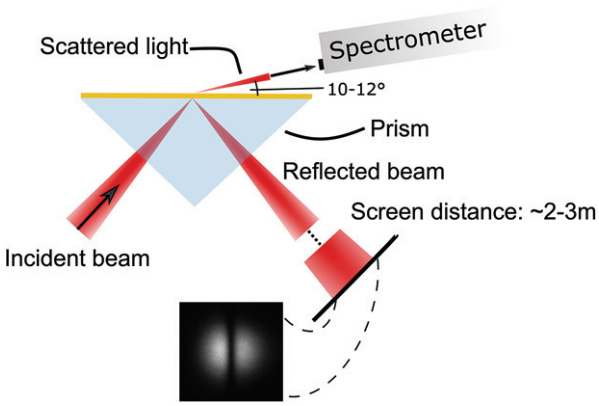

Fig. 1: (Colour online) The layout of the experimental setup. The SPPs were generated in the Kretschmann configuration on thin gold layers (with 45 nm thickness). The position of resonance was measured by imaging the reflected beam to a CCD camera being far from the location of the gold surface. The spectral distribution of the SPP emitted light was detected at the optimal emission angle [17] and coupled into the spectrometer via an optical fiber.

The angle of the incident beam is $\alpha = 44.9$, the amplitude of the measured angular shift oscillation is about $\delta\alpha \approx 4.1\ 10^{-4}$ (deg) $= 7.2\ 10^{-6}$ (radian).

When looking for potential explanations of this effect, attributed to the periodically changing dispersion as a function of laser intensity we refer to the works of Zhang [18], finding similar effects in the terahertz

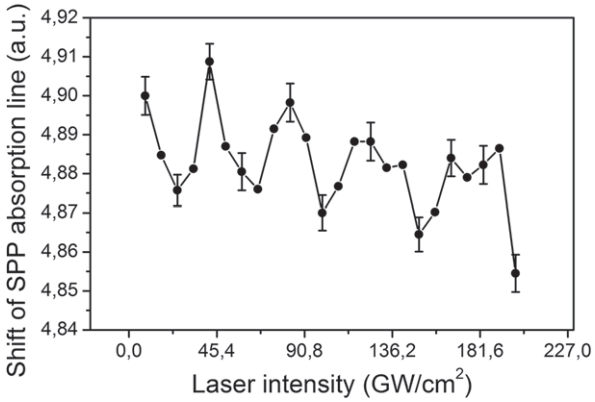

Fig. 2: Laser intensity dependence of the angular minimum in the reflected beam. The SPPs were excited by missing photons. (in the geometry of fig. 1). The minimum position has been determined by fitting a Gaussian curve to the distribution around this minimum. The exciting laser beam has been focused to the gold film via a glass prism in the Kretschmann geometry.

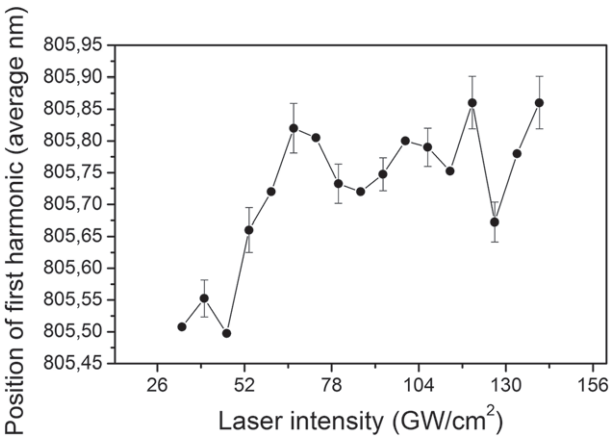

Fig. 3: The laser intensity dependence of the central wavelength of the basic harmonic light emitted by decaying SPPs. The wavelength values have been obtained by fitting Gaussian curve to the measured spectral line. The oscillatory change is below 0.1%, indicating that in the given exciting laser intensity range there is no significant change in the position of this spectral line.

frequency range. His data show also periodicity, but this effect is smoothing out at higher frequencies. At the same time this paper predicts the decrease of the energy/frequency of plasmonic dispersion. In [18] the oscillatory character of the change of dispersion is attributed to the dynamical screening effect of electrons in the strong field.

In order to make the situation clear in our case, where the frequency of the exciting laser is almost three orders of magnitude higher than in [18], the spectral properties of the SPP emitted light have been studied. The layout of the experimental facility is also shown in fig. 1. The result of the average of several runs is shown in fig. 3. Here the data coming from Gaussian fits to the spectral distribution of the basic harmonic of the SPP emitted light is plotted as a function of laser intensity. It is seen that there is no significant change in the dispersion properties, the laser intensity dependence of the spectral shift is less than 0.1%. It is also seen in the figure that the laser intensity dependence of the spectral position is similar to that of fig. 2. In all our measurements the laser intensity has also covered the 20 to 200 GW/cm^2 intensity range and in all cases of the spectral measurements the oscillatory behavior could be detected.

In order to find some theoretical explanation of these findings efforts were made to incorporate the influence of dynamical screening into the dielectric function and through it into the dispersion properties of our gold film. The measured dispersion looks to be dependent on laser intensity, as it should if the process behind it is dynamical screening. This has been the motivation to perform calculations in this direction.

Let ε_1 denote the relative dielectric function of the metal depending on the momentum and energy, and ε_2 the relative dielectric function of the insulator or vacuum. Than the dispersion of SPP excited in the Kretschmann geometry (in the x-direction) is given by

$$q_x = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1+\varepsilon_2}}. \tag{1}$$

In our case, on the air side of the gold film $\varepsilon_2 = 1$. We are interested in the modification of the dispersion at the frequency of the incoming light as a function of its intensity only. As by our experimental results this is very small, we use the following simple picture to interpret the measurements. The dielectric function for a metal in RPA approximation

$$\varepsilon_1(q,\omega) = 1 + V_q \Pi^0(q,\omega), \tag{2}$$

where V_q is the Coulomb interaction and $\Pi^0(q,\omega)$ is the Lindhard function. For low light intensity, as $\hbar\omega \gg v_F q$, where v_F is the Fermi velocity,

$$\varepsilon_1(q,\omega) \approx \varepsilon_1(\omega) = 1 - \frac{\omega_{pl}^2}{\omega^2}, \tag{3}$$

where ω_{pl} is the bulk plasma frequency of the metal. Inserting this into (1), the usual SPP dispersion is recovered.

However at high light intensities, the electron states are heavily dressed by photons [18]. The electron wave functions instead of plane waves, when the vector potential for the laser field

$$A = e_x \frac{E}{\omega}\sin\omega t \tag{4}$$

will have the form

$$\psi_k^{(0)}(r,t) = e^{iF(t)} e^{i\gamma_0 k_x(1-\cos\omega t)} e^{ikr} e^{i\epsilon_k t}, \tag{5}$$

where $\gamma_0 = eE/(m^*\omega^2)$, $\gamma_1 = (eE)^2/(8m^*\omega^3)$, $F(t) = 2\gamma_1\omega t + \gamma_1 \sin 2\omega t$, and $\epsilon_k = \hbar^2 k^2/2m^*$.

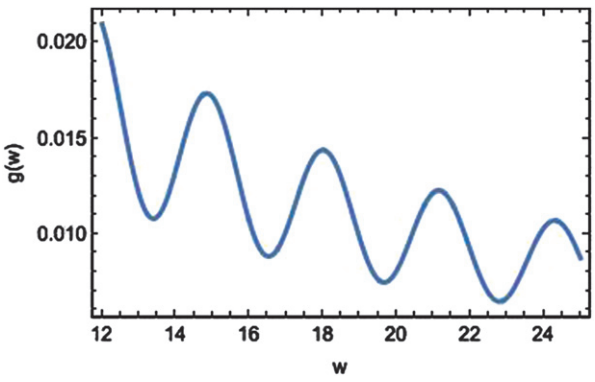

Fig. 4: (Colour online) The dimensionless electromagnetic-field dependence of the oscillatory screened part $g(w)$ (eq. (9)) of the dielectric function.

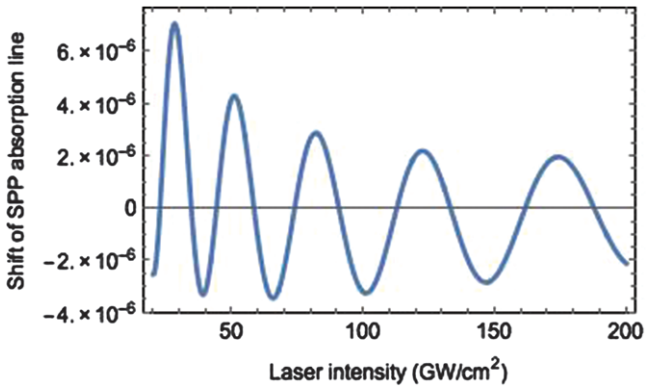

Fig. 5: (Colour online) The laser intensity dependence of the shift of the SPP absorption line, with $\beta = 0.013$.

Using this wave function, the polarization function instead of the Lindhard function, in the same RPA approximation

$$\Pi\left(q,\varpi\right)=\sum_{m}J_{m}^{2}\left(q_{x}\gamma_{0}\right)\Pi^{0}\left(q,\varpi+m\omega\right), \tag{6}$$

where $J_m(x)$ are the Bessel functions. In our case we are interested in the dielectric function at the frequency of the incoming light only, $\varpi = \omega$, therefore the polarization function

$$\Pi\left(\omega\right)\approx-\frac{\omega_{pl}^{2}}{\omega^{2}}\sum_{m}\frac{J_{m}^{2}\left(w\right)}{\left(1+m\right)^{2}}, \tag{7}$$

where $w = (cq_x/\omega)\, eE/cm^*\omega$.

Using this polarization function alone would lead to an unphysical result at high electric fields in this RPA approximation.

However, the intensive laser field is present only in a very small fraction of the sample ($\sim$20 μm diameter), therefore the response can be approximated by the sum of the response of the electrons which are in the strong field region, with weight β and the response of the electrons from the rest of the sample, where the electric field is negligible, with weight $(1-\beta)$, therefore the total dielectric function can be approximated by

$$\varepsilon_{1}\left(q_{x},\omega\right)\approx1-\left(1-\beta\right)\frac{\omega_{pl}^{2}}{\omega^{2}}-\beta\frac{\omega_{pl}^{2}}{\omega^{2}}g\left(w\right), \tag{8}$$

where

$$g\left(w\right)=\sum_{m}\frac{J_{m}^{2}\left(w\right)}{\left(1+m\right)^{2}}. \tag{9}$$

The oscillating part of the dielectric function is shown in fig. 4.

In the experimentally measured intensity region this $g(w)$ function has minima at $w \approx 13.31$, 16.46, 19.61, 22.75.

From the experimental data (fig. 2), assuming that $m^* = m$, the free electron mass, the values of w are approximately 13.85, 17.46, 21.67, and 25.43.

Comparing the theoretical data with the experimental ones, we find that the effective mass of the electrons is smaller than the free electron mass in the high plasmonic field and slightly decreases with increasing laser intensity, $m^*/m \approx 0.96, 0.94, 0.9, 0.89$, respectively. There is a surprisingly good agreement of the above-described simple model with our experimental data. In order to explain the found decrease of the effective mass with increasing laser intensity however, further investigations are needed.

Since we are in a momentum-frequency range, where the dispersion is nearly linear and the measured laser-intensity–dependent oscillation is very small, we may approximate in the argument of the Bessel functions $cq_x \approx \omega$ therefore $w \approx eE/cm^*\omega$. Inserting (8) into (1), we can see that the frequency of the oscillation of q_x depends on the strengths of the laser electric field, and its amplitude can be fitted by β.

Here we are interested only in the oscillatory part of the dispersion as a function of the laser intensity.

As $\beta \ll 1$, expanding the SPP dispersion, keeping only the first order in β, furthermore since $\omega/\omega_{pl} = 0.17$, keeping only the terms proportional to $(\omega/\omega_{pl})^2$, the angular shift of the plasmon resonance angle

$$\delta\alpha\approx\beta\frac{g(w)}{2}\frac{\omega^{2}}{\omega_{pl}^{2}}. \tag{10}$$

At the EM field frequencies used in our measurements, even without the laser field the SPP dispersion deviates from the linear mode, *i.e.* q_x is shifted, though in a very small amount, to larger values. In addition there is a further shift of the order of $\sim$0.1% due to the intensive laser field. Neglecting these shifts, keeping only the oscillatory parts in δq_x, with $\beta = 0.013$, the laser intensity dependence of $\delta\alpha$ is shown in fig. 5, being in good agreement with the experimental data of fig. 2.

Conclusions. – The SPP exciting laser intensity dependence of the properties of the SPP emitted photons has a weak oscillatory part in the measured 20–200 GW/cm^2 intensity range. This indicates a dynamic screening effect.

A simple model of dynamic screening describes the experimental observations reasonably well if, in addition to the dielectric function for the about 20 micron diameter exciting laser spot, that of the surrounding, not excited part of the gold film is taken into account. This can be justified by the long range of the Coulomb interaction. The effective mass of the electrons is smaller than that of free electrons due to the screening effect and decreases with increasing laser intensity.

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REFERENCES

[1] Kauranen M. and Zayats A. V., *Nat. Photon.*, **6** (2012) 737.
[2] Foley J. J. IV, Harutyunyan H., Rosenmann D., Divan R., Wiederrecht G. P. and Gray S. K., *Sci. Rep.*, **5** (2015) 9929.
[3] Grosse N. B., Heckmann J. and Woggon U., *Phys. Rev. Lett.*, **108** (2012) 136802.
[4] Park I.-Y. *et al.*, *Nat. Photon.*, **5** (2011) 677.
[5] Rotenberg N., Betz M. and van Driel H. M., *Phys. Rev. Lett.*, **105** (2010) 017402.
[6] Krasavin A. V., MacDonald K. F., Zheludev N. I. and Zayats A. V., *Appl. Phys. Lett.*, **85** (2004) 3369.
[7] Rotenberg N., Caspers J. N. and van Driel H. M., *Phys. Rev. B*, **80** (2009) 245420.
[8] Kroó N., Farkas G., Dombi P. and Varró S., *Opt. Express*, **16** (2008) 21656.
[9] Brongersma M. L., Halas N. J. and Nordlander P., *Nat. Nanotechnol.*, **10** (2014) 25.
[10] Yun L., Qiang L., Shimeng C., Fang C., Hanqi W. and Wei P., *Sci. Rep.*, **5** (2012) 12864.
[11] Nielsen M. and Elezzabi A. Y., *Opt. Express*, **20** (2012) 8592.
[12] Ayala-Orozco C. *et al.*, *ACS Nano*, **8** (2014) 6372.
[13] Dombi P. and Antal P., *Laser Phys. Lett.*, **4** (2007) 538.
[14] Kroó N., Rácz P. and Varró S., *EPL*, **105** (2014) 67003.
[15] Kroó N., Rácz P. and Varró S., *EPL*, **110** (2015) 67008.
[16] Kroó N., Rácz P. and Varró S., *Nucl. Instrum. Methods B*, **369** (2016) 55.
[17] Kroó N., Varró S., Farkas G., Dombi P., Oszetzky D., Nagy A. and Czitrovszky A., *J. Mod. Opt.*, **55** (2008) 3203.
[18] Zhang C., *Phys Rev. B*, **65** (2002) 153107.