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Bright new world

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ABSTRACT

Surface plasmons (SPOs) have been excited by intense femtosecond laser pulses on a gold film at room temperature and their near field has been analyzed by the intensity dependent response of an STM and by studying the spectra of multiplasmon emitted electrons. Around 80 GW/cm^2 laser intensity, anomalies have been found in both cases, interpreted as the stepping in of electron pairing, transition to a diamagnetic state, and by anomalous Faraday rotation.

1. Introduction

Electromagnetic radiations are parts of our everyday life. The near UV–visible–near IR range, practically represented by sunshine is treated by a special branch of science called optics. The invention and development of lasers and their broad field of applications in various technologies have aroused increasing attention in many societies. One clear signal of this phenomenon is the International Year of Light 2015 and its enthusiastic global acceptance.

In line with the recent developments in nanoscience and nanotechnology, nanooptics is also in the centre of scientific and technological interest. Therefore, the most significant hurdle in optics, namely the existence of a diffraction limit cries out for new solutions, first of all in the fields of nanoscience and nanotechnology. Plasmonics is one, perhaps the most important option to bypass this hurdle, since the diffraction limit can be overcome and, in addition, electromagnetic radiation energy can be squeezed into small, even nanoscale volumes, resulting in extremely high EM fields. Since surface plasmons, the quasi particles of plasmonics, can be excited efficiently with near UV–visible–near IR light, relatively low laser intensities can be used to study and explore high field effects, e.g., nonlinear optical phenomena even on nanoscale.

2. Experiments

In one of our recent work we studied the light emission from smooth Au films when the SPO-s were excited by intense fs pulses of a Ti:Sa laser. Here we have also found light amplification [1] and line narrowing above a few ten GW/cm^2 laser intensity as shown in Fig. 1 again implying laser like actions.

But at the same time, if laser intensity has been sufficiently high, multiplasmon electron emission could also be induced at orders of magnitude lower laser intensity than, e.g., in gases. Some of the first experiments of this type were performed on atoms in gas phase [2]. These atoms can be ionized in the high fields, the intensity being in the order of 10^{15} W/cm^2. In the case of metal surfaces this threshold can be orders of magnitude lower, due to the involvement of SPO-s in the process [3]. If these quasi-particles are excited resonantly on the metal (e.g., Au) surface, the electromagnetic energy is squeezed at the surface and laser intensities in the order of 10^{10} W/cm^2 are already inducing a high intensity of SPO-s, leading to multiplasmon decay into hot electrons with energy above the work function of the metal. In these cases electron emission from the surface can be observed. A typical electron emission–laser intensity plot for a gold film is shown in Fig. 2 in double logarithmic scale [4].

The slope of ~4 indicates a four photon (SPO) process arriving at saturation around 80 GW/cm^2 in agreement with previous measurements [3] interpreted on the basis of the Keldysh theory [5], explaining the "saturation" by the stepping in of tunneling of the emitted electrons. It is worth remembering here that the issue of hot electrons has become a hot topic recently [6] because of the interesting science behind it and the numerous potential applications [7].

If the spectrum of emitted electrons is analyzed by the time-of-flight method it is found that at around 80 GW/cm^2 laser intensity a separate high energy peak shows up in a relatively broad laser intensity range (Fig. 3) which also increases with the increase of exciting laser intensity, but does not saturate around 80 GW/cm^2,

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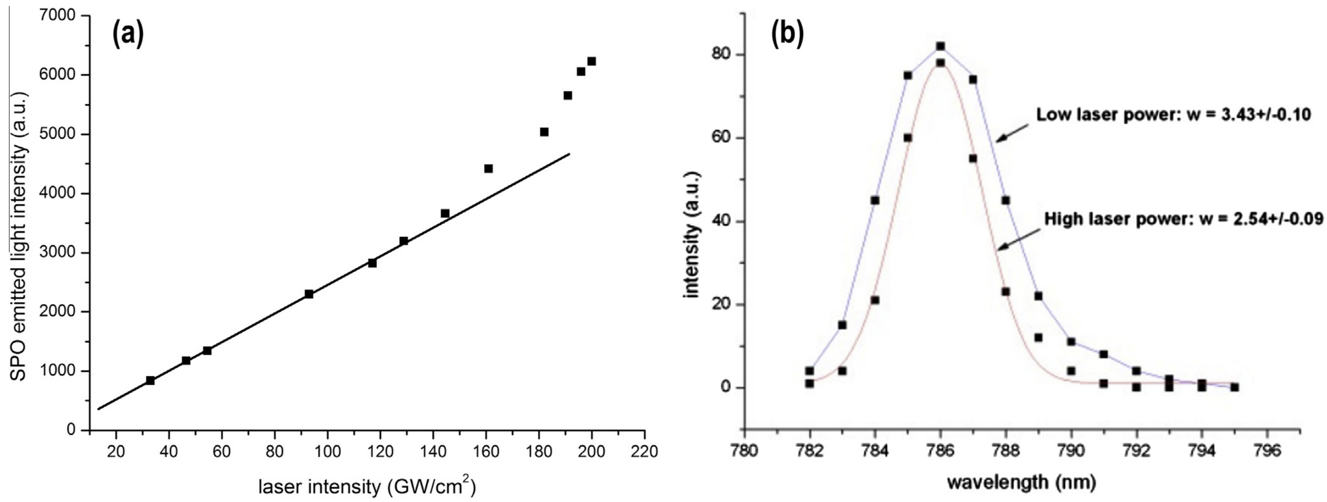

Fig. 1. (a) Laser intensity dependence of light intensity, emitted by decaying SPO-s and (b) the laser intensity dependent spectral narrowing of this emitted light.

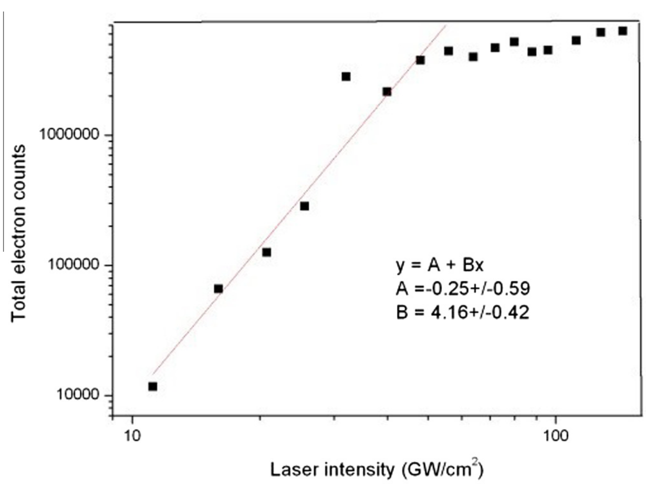

Fig. 2. Laser intensity dependence of the total number of emitted electrons.

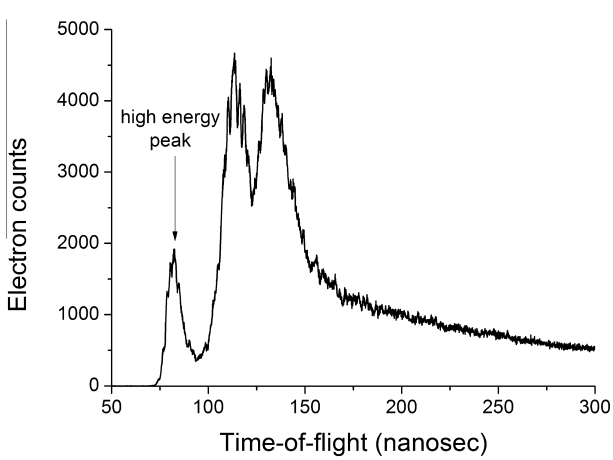

Fig. 3. Time-of-flight spectrum of multiplasmon emitted electrons with a well pronounced high energy peak attributed to electron pairing around 80 GW/cm^2 laser intensity.

rather starts to decrease as it has been shown by us in [8] and presented here in Fig. 4. The remaining part (>95%) of the spectrum has the intensity dependence of Fig. 2.

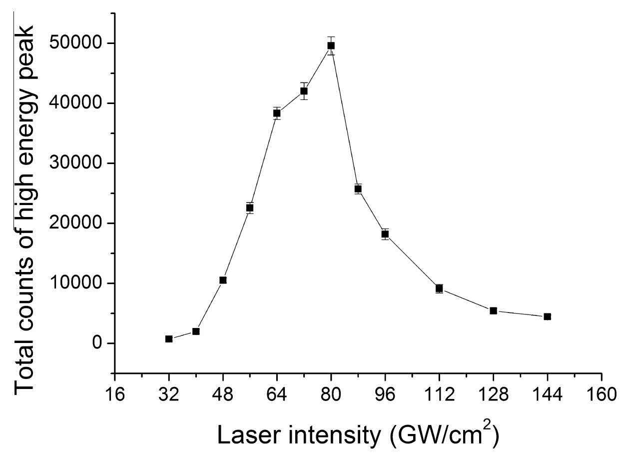

Fig. 4. Laser intensity dependence of the high energy peak of Fig. 3.

This effect has been interpreted as the result of the pairing of electrons on the metal surface at room temperature, being at a few tens of nanometers distance from each other [9].

Surface plasmons are influenced by the topography of the surface, they may get localized. These localized SPO-s have also many special features. One of them is that they squeeze light into small, even nano-sized structures. This may lead to the hosting of extremely high EM fields, called hot spots. These hot spots are the sites of a large number of hot electrons. Near field microscopes, like SPO near field STM-s, are ideal instruments to study the properties of these LSPO-s. We used this type of microscope to study SPO-s on gold surfaces in general and hot spots on this surface in particular. A typical pair of topological and SPO images is shown in Fig. 5 and the detailed study of the field enhancement on the surface is described in [10]. The STM rectifies the high frequency EM field and this leads to an SPO signal. This means that in hot spots STM signals are measured even if the STM bias is zero (Fig. 6).

When we studied our STM cases, where the SPO-s were excited with intense fs Ti:Sa laser pulses, we found similar anomaly in the response time of the STM to that of the high energy peak of the TOF spectrum of electron emission [8]. Some data are shown in Fig. 7 indicating that the lifetime of the observed anomaly (electron pairing lasts for about 7 orders of magnitude longer than that of the SPO exciting ~100 fs laser pulse. The anomaly found in both the TOF and STM cases could be interpreted on the basis of the analogy