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2003 J. Phys.: Condens. Matter 15 L349

(http://iopscience.iop.org/0953-8984/15/23/102)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) L349–L352 PII: S0953-8984(03)62488-4

**LETTER TO THE EDITOR**

# Strong second-harmonic radiation from a thin silver film with randomly distributed small holes

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Received 23 April 2003
Published 30 May 2003
Online at stacks.iop.org/JPhysCM/15/L349

**Abstract**
We report the observation of strong second-harmonic radiation from a thin silver film containing randomly distributed small holes. A pulsed laser beam of wavelength 1064 nm impinges at an angle of incidence 45° on the film, and the reflection is collected by a CCD detector and analysed by a high-resolution spectrometer. Strong second-harmonic radiation was observed at the wavelength of 532 nm with a halfwidth of 40 nm.

Silver thin films have been intensively studied, particularly as regards the phenomena related to the surface plasmon excitation [1, 2]. Second-harmonic generation from thin silver films has also been studied for a long time [3–8]. The second-harmonic generation can be dramatically enhanced via the surface roughness [6] or the resonant transitions of bulk and surface states [7].

A recent discovery in the optics of thin metallic films is the so-called extraordinary optical transmission through subwavelength hole arrays in a thin silver film [9–11]. This enhancement is huge (up to a factor of 2), considering the theoretic prediction ($\sim(r/\lambda)^4$, where $r$ is the radius of the hole) based on the well-known Bethe theory [12]. It is believed that the excitation of the surface plasmon by the grating on the surface is responsible for the extraordinary enhancement of the transmission through the subwavelength holes. More recently, it was however demonstrated theoretically [13] that the subwavelength holes were not necessary for the resonant transmission. And we have also demonstrated experimentally [14] that while the small holes induce a strong near-field transmission, the holes are not very helpful for the far-field transmission. The strong near-field transmission indicates that surface plasmon polaritons are established on the two sides of the film [14]. We have also discovered [15] that the established surface waves can travel along the film for a long distance with a spectrum red-shifted some 200 nm. In our experiments [14, 15] the holes were randomly distributed and the size and shape of the holes vary too. The lack of periodic objects in the samples suggested

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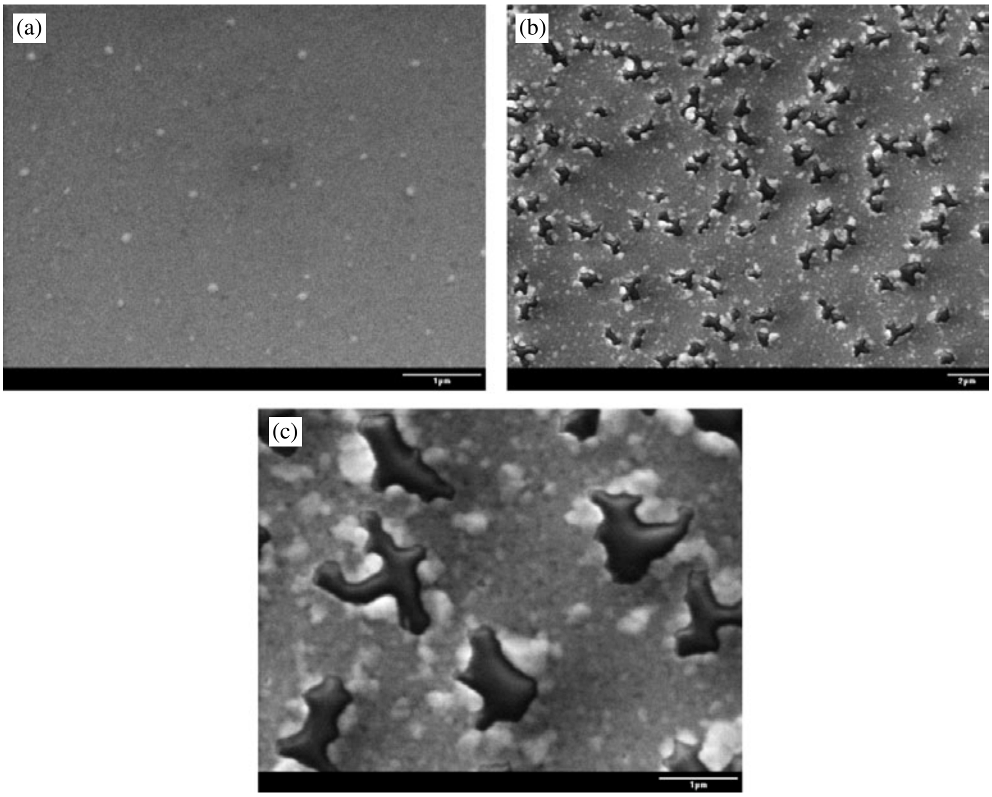


**Figure 1.** SEM images for the silver thin film (a) without holes; (b) with holes; (c) enlarged from (b).

that the induced strong near-field transmission would be generated by strong coupling of the surface plasmon on two sides of the film [14].

Second-harmonic generation from a thin silver film with a small hole embedded in a spherical grating was recently observed [16]. In the present work we have measured nonlinear responses from a randomly porous film. We have observed strong second-harmonic radiation from the film.

The sample was a silver film of thickness 60 nm deposited on glass substrate (Corning, Micro Slides, 1 mm thickness). We used a simple method to produce the sample [17]. First, we prepared a thin silver film of thickness 60 nm by the usual vapour deposition process. We then applied a DC voltage upon the film. The film was monitored under an atomic force microscope. After some time, the film started to show microscopic holes. We stopped the process when we saw the image we wanted to study. We have carried out scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) on the film. In figure 1 SEM images of the prepared film are presented. The SEM study shows the following. Uniformly in the film, there are randomly distributed small holes. The size of the holes ranges from 100 to 1000 nm, and the shape of the holes varies dramatically. The distance between the holes varies from 200 to 2000 nm. For comparison, a SEM image of the film without holes is also presented in figure 1.

It is interesting to note the following. In [9–11] the samples contained only a few small holes that have a unique size and shape and are distributed with a unique distance between the

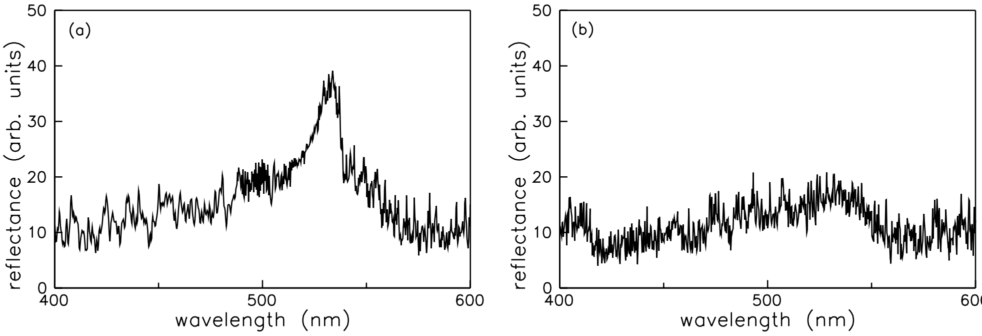


**Figure 2.** Reflectances from the silver thin film (a) with holes and (b) without holes. The two figures have the same scale.

holes, so the film acts as a grating. Our samples are different. The main differences are as follows. Macroscopically, the small holes are uniformly distributed everywhere in the whole sample area. Microscopically, the distances between the small holes are random and the size as well as the shape of the holes vary too. In other words, there are not the uniform objects and unique periodicity required for the system to be considered as a grating structure.

Measurements of nonlinear responses were made on the prepared samples, with a $Q$-switch Nd:YAG pulsed laser (Spectra Physics, wavelength 1064 nm, repeating frequency 10 Hz and pulse duration 10 ns). The pulse energy was limited to less than 4 mJ/pulse in a 4 mm diameter spot, so as to avoid any damage to the samples. The angle of incidence was chosen to be 45°, and the reflection was collected by a CCD detector and analysed by a high-resolution spectrometer (CVI, SMGT-2). In the following we shall present some spectra recorded from the experiments.

In figure 2 there are two spectra. The spectrum In figure 2(a) was obtained from the film with holes and that in figure 2(b) from the film without holes. The scales of the reflection ($y$-axis) in the two figures (figures 2(a) and (b)) are the same, so the two spectra can be compared directly. One can readily see in figure 2 the second-harmonic generation at wavelength 532 nm, especially for the film with holes. For the film without holes the second-harmonic generation can hardly be seen above the background noise, whereas for the film with holes the second-harmonic generation appears clear and strong. We have measured the halfwidth of the second-harmonic generation peak in figure 2(a) by enlarging and smoothing the curve near the peak. It turns out that the second-harmonic generation is a Gaussian pulse and has a halfwidth of about 40 nm.

It is widely believed that the strong second-harmonic generation on rough silver thin films is due to the resonant or near-resonant local field enhancements [4]. Actually the enhancement includes two steps. The fundamental field can be self-consistently enhanced due to the microscopic structures on the surfaces of the film [18], and subsequently the excited second-harmonic generation is again enhanced due to the same strongly interacting microscopic structures. In the process, strong surface waves, surface plasmon polaritons, can be established on the two surfaces of the film. On a smooth surface the surface waves are highly localized on the surface. The waves can radiate into the space if the surface is rough and contains microscopic structures. In our present work, the subwavelength holes in the film have several effects as follows. The holes bear strong interactions between the surface plasmons on two sides of the film [14]. The holes generate strong local field enhancements [18]. The holes provide the two-dimensional propagation along the surface [15]. And the surface waves radiate into the space because of the holes.

It is important to emphasize that in the present work the samples had no grating structures. It is possible that grating structures played a critical role in the linear transmission reported in [9] as well as in the nonlinear responses presented in [16]. The present work suggests however that the enhanced second-harmonic generation via small holes in a thin silver film may occur without a grating structure. That is, the holes can be just randomly distributed in the film and the size and shape of the holes can be arbitrary too.

On the basis of the results presented in the present communication and from previous work in [15] and [16], one is led to conclude additionally that the grating structures are not essential for the linear transmission or for the enhanced second-harmonic generation in reflection to occur.

In conclusion, we have reported the observation of some nonlinear optical responses from a thin silver film containing randomly distributed small holes. The strong radiation at a half of the fundamental wavelength is identified as the parametric second-harmonic generation. This is a simple, inexpensive and reliable way to acquire pulsed light source at this frequency.

We thank N Perea-López and I Gradilla for assistances in the experiments. The work was supported by the Consejo Nacional de Ciencias y Tecnología (CONACyT-México), Project No 32268-E and the Direccion General de Asuntos del Personal Académico, Universidad Nacional Autónoma de México (DGAPA-UNAM), Project No IN102600.

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