Detection of Transient Reflection Gratings under Surface Plasmon Resonance Condition: Numerical Simulation

Yao-chun Shen[*1,2], **Zuhong Lu**[1] and **Shu-yi Zhang**[2]

[1]*National Laboratory of Molecular and Biomolecular Electronics, Southeast University, Nanjing 210096, China*
[2]*National Laboratory of Modern Acoustics, Nanjing University, Nanjing 210093, China*

Based on rigorous electromagnetic wave theory, a theoretical analysis is presented to study the laser beam reflected and diffracted from the metallic grating, which is excited in the surface plane by interference of two pulsed laser beams. Numerical results show that, at some distinct incidence angles surface plasmon resonance occurs and the intensity change of reflected light is about 50 times larger than that of diffracted laser beam under normal incidence. This suggests a sensitive method for detecting transient reflecting gratings, especially in real time measurements.

The laser-induced transient grating technique is a useful non-contact and non-destructive method for studying a wide variety of material properties, particularly the thermal and acoustic properties of thin films and interfaces.[1,2] In experiment, two temporally coincident laser pulses (pump laser pulse) are crossed at the sample surface. Absorption of crossed laser pulses in a sample gives rise to a spatially periodic heating and thermal expansion, which launches counter-propagating acoustic waves. In almost all transient grating experiments, the acoustic oscillations and their delay, and finally the thermal diffusion, are usually monitored through the measurement of the time-dependent diffraction of a third laser beam (probe laser pulse), and provide information about the thermal and elastic properties of thin film and bulk materials.[3-6] The laser intensity diffracted from a metallic grating is proportional to the square of the grating amplitude, which is usually very small and determined by the intensity of the pump laser pulse. In order to increase the signal-to-noise ratio the date acquisition involves many repetitions of a pump-probe sequence. This leads to long date collection times and in some case to cumulative sample heating and damage. Recently, real-time measurements have been achieved by using a cw laser or "quasi-cw" laser pulse as probe beam.[7,8]

It was shown that when a laser beam is incident on a metallic grating with a distinct incident angle, resonance absorption occurs due to the excitation of surface plasmon, which is a kind of surface-localized plasma wave propagating along a metal/dielectric interface.[9] The surface plasmon is very sensitive to the changes of interface properties due to the fact that its energy is concentrated within the evanescent field near the interface. Very recently Katayama et al.[10] reported the laser generation of transient reflecting gratings (TRG) under surface plasmon resonance (SPR) condition. In this paper we will show that by monitoring TRG under SPR condition, a much higher detection sensitivity should be obtained.

Outline of the theory

When a probe laser beam incidents on the surface of a metallic grating with an incidence angle θ, light will be reflected and
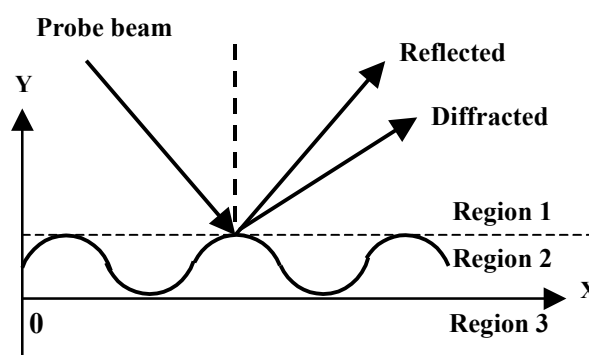
* Corresponding author: ycshen@seu.edu.cn

Fig. 1 Schematic representation of light diffraction from a metallic grating surface.

diffracted at the surface of the grating as shown in figure 1. For simplicity, we assume that the incidence probe beam is a p-polarized HeNe laser. Thus the z-component magnetic field, which is the only magnetic component for p-polarized plane wave, can be expressed as following Rayleigh expansion outside the modulated zone (region 1 and 3),[11]

$$H_{z1}(x,y) = \sum_{n=-\infty}^{\infty} \left(A_n e^{-j\beta_{n1}y} + B_n e^{-j\beta_{n1}y} \right) \cdot e^{j\alpha_n x}$$

$$H_{z3}(x,y) = \sum_{n=-\infty}^{\infty} C_n e^{-j\beta_{n3}y} \cdot \exp(j\alpha_n x)$$

and inside the modulated zone (region 2) it can only be expressed as Fourier expansion:

$$H_{z2} = \sum_{n=-\infty}^{\infty} H_n(y) \cdot e^{j\alpha_n x}$$

where $\alpha_n = k_0 \sin\theta + 2n\pi/d$, $\beta_{ni}^2 = \varepsilon_i k_0^2 - \alpha_n^2$,

d is the grating period, k_0 and ε_i is the wave vector of light in vacuum and the complex dielectric constant of the i-th region, respectively. Note that the dielectric constant of region 2 is a function of the position (x,y), general the Fourier expansion coefficient $H_n(y)$ has no analytical expression. This problem can be solved by numerical calculation using finite element method.[11] The Rayleigh expansion coefficients (A_n, B_n, C_n) can then be determined through the boundary conditions at y=0 and y=2h, where h is the grating depth.

Once the Rayleigh and Fourier expansion coefficients are determined, the intensity of light reflected and diffracted from the metallic grating can easily be calculated. In all numerical calculations the wavelength of He-Ne laser (632.8 nm) and the dielectric constant of silver (-17.4, 0.56) are used.

Results and discussions

Validity of the theoretical model

Figure 2 shows the calculated optical absorption curve as a function of incidence angle for a silver grating with a grating period of 2000 nm and a grating depth of 100 nm. In the absorption curve there are three absorption peaks at 4.6°, 23.4°, 45.5°, which are explained as due to the third-, second- and first-order SPR mode of the silver gratings. It was shown that the peak position of the optical absorption curve due to surface plasmon on a metallic grating can be described by perturbation theory as $\sin\theta = (\varepsilon/(\varepsilon'+1))^{1/2} \pm n\lambda/d$.[9] By using the real part of the complex dielectric constant of silver at HeNe wavelength, the peak positions were calculated to be 45.56° for n=1, 23.43° for n=2 and 4.66° for n=3, which are in very good agreement with our numerical results based on finite element method. Note that the perturbation theory can provide the correct peak position, while the finite element method can be used to calculate the peak positions, the intensity and the shape of the absorption curve as well.

Fig.2 also shows the experimental result measured previously with photoacoustic method.[12] Obviously the calculated absorption curve is in very good agreement with that of experiment including the correct prediction of the small peak at 4.6°.
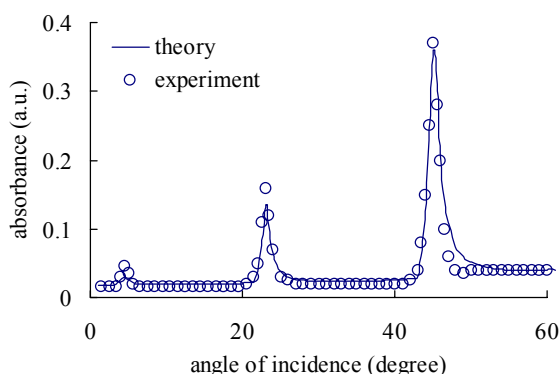
Fig. 2 The calculated and measured optical absorbance of a silver grating for p-polarized light as a function of incidence angle. The grating period is 2000 nm and the depth is 100 nm.

Numerical simulations

With the validity of our theoretical model, the possibility of laser detection of metallic grating under SPR conditions was studied. Figure 3 shows the calculated intensity of the reflected light from the surface of silver grating with a grating period of 1000 nm as a function of incidence angle. There is a sharp peak in the reflection curve. The intensity of the reflected light decreases dramatically with the increase of the grating depth at the incidence angle of 23.44°. It is interesting to note that the intensity of the reflected light reaches zero at a grating depth of 20 nm, indicating that there will be no reflection from the metallic reflecting grating surface at this special case. The inset of Fig.3 shows the intensity of the reflected light as a function of grating depth. It is clear that under normal incidence condition (θ=0°), the intensity of the reflected light decreases slowly as the increase of the grating depth. However, at the incidence angle of 23.44°, the intensity of the reflected light decreases dramatically with the increase of grating depth. This is due to the resonance excitation and absorption of surface plasmon.
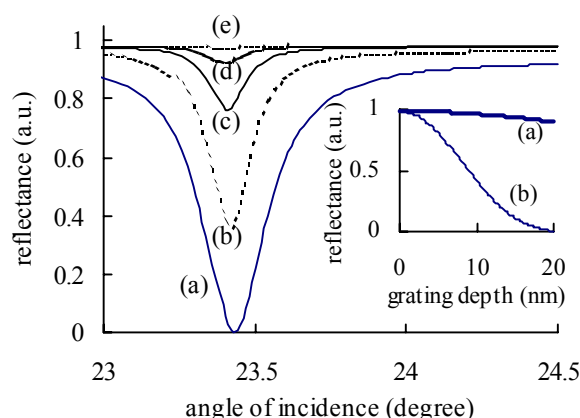
Fig. 3 Intensity of the reflected light from a silver grating changes with incident angle, the grating period is 1.0 mm and the grating depth is 20.0 nm (a), 10.0 nm (b), 5.0 nm (c), 2.5 nm (d) and 1.0 nm (e), respectively. The inset shows the intensity of the reflected light versus grating depth under normal incidence (a) and for an incidence angle of 23.44°.

Curve (a) in Fig. 4 shows the intensity of the first-order diffracted light from a silver grating under normal incidence, which is used to monitor transient grating in TRG experiments.[2,7] The slope of this curve is 0.5, which is reasonable since the first-order diffraction efficiency from metallic grating of small depth is proportional to the square of the grating depth.[9] Curve (b) in Fig.4 represents the change of laser intensity reflected from the same grating under surface plasmon resonance condition (angle of incidence 23.44°). It is obvious that the change of reflectivity, defined as the difference of the reflectivity between a smooth silver surface and that of a grating surface, is about 50 times larger than the intensity change of the first-order diffracted beam under normal incidence. This implies that by monitoring the intensity change of the reflected light, instead of monitoring the intensity of the diffracted light, a much high sensitivity could be expected. Note that the first-order diffracted laser beam becomes evanescent mode due to surface plasmon resonance, and the intensity of the diffracted laser beam approaches zero at this specific incident angle of 23.44°.

In a typical TRG experiment, the transient gating is monitored through the measurement of the diffraction of a probe beam. Usually the probe beam is also a laser pulse and the time dependence of the response is obtained by repeating
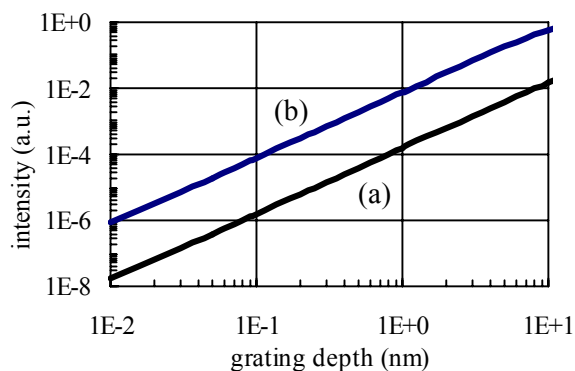
Fig. 4 Numerical simulation for the laser detection of LITGs, (a) Intensity of the first-order diffracted light from a silver grating under normal incident, the grating period is 1000 nm. (b) Intensity change of the light reflected from the same grating for an incidence angle of 23.44°. Note that (b) is 50 times larger than (a).

the pump-probe pulse sequence with different delays of the probe relative to the pump. This approach provides very high sensitivity since the power intensity of the laser pulse diffracted from the grating is large. It has been shown that by using a cw laser, instead of pulsed laser, as the probe beam, real-time detection of TRG has been achieved.[7,8] The real-time detection scheme allows the entire response to be measured with minimal signal averaging and even a single laser pulse can yields an adequate signal. The sensitivity of the real-time measurements is determined by the power intensity of the cw probe laser. The numerical results presented in this paper shows that the sensitivity could be greatly improved by monitoring the intensity change of the reflected light, instead of monitoring the intensity of the diffracted light under normal incidence.

In conclusion, a finite element method based on rigorous electromagnetic wave theory is presented to study the intensity change of the laser beam reflected and diffracted from silver gratings. Numerical results show that, at some distinct incidence angles surface plasmon resonance occurs and the intensity change of reflected light is about 50 times larger than that of diffracted laser beam under normal incidence. This suggests a sensitive method for detecting transient reflecting gratings, especially in real-time measurement.

References:
1.   K. A. Nelson, R. Casalengno, R. J. D.Miller and M. D. Fayer, *J. Chem. Phys.*, **1982**, 77, 1144
2.   A. Harata, H. Nishimura and T. Sawada, *Appl. Phys. Lett.*, **1990**, 57, 132
3.   A. R. Doggal, J. A. Rogers and K. A. Nelson, *J. Appl. Phys.*, **1992**, 72, 2823
4.   A. Harata and T. Sawada, *Trends Anal. Chem.*, **1995**, 14, 504
5.   Q.Shen, A.Harata and T.Sawada, *J. Appl. Phys.*, **1995**, 77, 1488
6.   Q Shen, A. Harata and T. Sawada, *Jpn. J. Appl. Phys.*, **1996**, 35, 2339
7.   J. A. Rogers and K. A. Nelson, *J. Appl. Phys.*, **1994**, 75, 1534
8.   Y .C. Shen and P. Hess, *J. Appl. Phys.*, **1997**, 82, 4758
9.   H. Raether, *Surface plasmons on smooth and rough surfaces and on gratings*, Springer Tract Mod. Phys., Springer, Berlin, Heidelberg, **1988**, 111, 91
10.   K. Katayama, Q. Shen, A. Harata and T. Sawada, *Appl. Phys. Lett.*, **1996**, 69, 2468
11.   P. Vincent, in *Electromagnetic Theory of Gratings*, ed. R. Petit, *Topic Curr. Phys.*, Springer,Berlin, Heidelberg, **1980**, 22, 101
12.   Y. C. Shen, S. Y. Zhang, Y. B. Zheng, *Science in China*, **1994**, A24, 967 (in Chinese)