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Thickness dependence of infrared reflectance of ultrathin metallic films: Influence of quantum confinement

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ABSTRACT

In this letter we discuss experimental results on optical reflectance of ultrathin metallic films. The laser light source was tuned at the infrared wavelength of $\lambda = 9.2\mu m$. Three metals were tested: aluminum, niobium, and nickel. The thin films were coated on various wafers, namely, fused glass, α-quartz and amorphous silicon, with the technique of magnetic field assisted sputtering modulated at radio frequencies. The infrared reflectance was recorded while the thickness of the film varied from 5 to little more than 100 Å. A phenomenon is observed, i.e. a periodic oscillation appeared modulated on the otherwise classic thickness dependence of metallic films. The effects are attributed to the influence of quantum confinement induced intraband redistribution of conduction electrons. A one-dimensional quantum well model was employed to simulate the system, and the numerical results confirmed the existence of the quantum size effect.

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1. Introduction

Optical responses of solid thin films have always been in research focuses for the physics as well as for various applications, such as packaging, anti-oxidizing, and electromagnetic isolation, etc. In addition, for ultrathin metallic films, i.e. the thickness falls around or below 10 nm, the optical properties of the films induced by quantum confinements may have impacting influences on fabrication and testing of various electric, optic, and opto-electric chips [1–4]. Despite the fact that optics of thin films have been well studied and relevant basic knowledge can be found in the literature, textbooks and scientific papers, ultrathin metallic films are generating new challenges from time to time due partially to the difficulties to fully understand some observed unusual phenomena. Among others, various quantum size effects are often the sources of the unusual phenomena [5–9].

One of the quantum size effects, intraband transition, will be explored in the present work, both experimentally and theoretically. In order for any quantum effects to be manifested significantly, there should be enough quantum confinements.

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For solids, quantum confinements of various types take place in molecular, grains, or aggregators. For metals, as far as the conduction electrons are concerned, strong quantum confinements may be established in some confined structures, such as, small metallic particles (often referred as quantum dots), small metallic rings (often referred as quantum rings), and ultrathin metallic films (often referred as quantum wells). The degree of the quantum confinements depends on the size and shape of the confined structures. In general, higher quantum confinements take place with smaller structures, i.e. the smaller object size the higher degree of quantum confinements. Due to the strong quantum confinements, the conduction electrons have to be at discrete energy levels. Therefore, the behavior and redistribution of the conduction electrons are subject to the quantum mechanics. To explore the system in details, one solves the eigen value problem defined by the time-independent Schrödinger equation plus the adequate boundary conditions. The solutions of eigen value problems are the eigen value, i.e. energy dispersions, and the eigen vectors, i.e. wave functions. The conduction electrons are redistributed among the discrete energies, according to Fermi-Dirac statistic distribution, and the locations or the probability distributions of the electrons are determined by the corresponding wave functions.

Obviously, the above described quantum confined objects will have properties and behaviors different from the classic unconfined materials. These various differences may be manifested, among other media, via light-mater interactions, and therefore demonstrate various quantum effects. The reasons for the quantum effects to occur can be in general divided into two classes, namely, the interband and intraband transitions. In interband transitions, when the inputting light is of high enough energy, electrons jump between discrete energy levels and produce paramagnetic radiations. The present work does not involve the paramagnetic effects as the light source we used is of low energy, i.e. in infrared range $\lambda = 9.2\mu m$. What being explored in the present work is the so-called diamagnetic effects which stem from the intraband transitions. In what follows in Section 3, the mechanism will be explained in more details.

In Section 2 of the paper, experimental results will be shown on the infrared optical reflectance of ultrathin metallic films. Three metals were tested, aluminum, nickel and niobium. The thin films were coated on various wafers, namely, fused glass, α-quartz and amorphous silicon, with the technique of magnetic field assisted sputtering modulated at radio frequencies. The infrared reflectance was recorded while the thickness of the film varied from 5 to little more than 100 Å. The experimental results are discussed and a periodic oscillation modulated on the thickness dependences is identified and considered stemming from the quantum size effects.

In Section 3 of the paper, a theoretic calculation based on one dimensional quantum well is carried out. The purpose of the calculation is to support the assertion made based on the experimental observation in Section 2. The time-independent Schrödinger equation is solved and both the energy levels and wave functions are determined. The conduction electrons are redistributed according to Fermi-Dirac statistic distribution and the electron density n is calculated by summing up all the electrons. Since the wave functions are different for different quantum well widths (the thickness of the films), an extra size dependence is introduced into the otherwise flat electron density. The size dependent electron density is used in Drude model to calculate the dielectric function ε and eventually the optical reflectance R. The results of the numerical simulation confirm the existence of the quantum size effect. The work is finally to be concluded in Section 4.

2. Experimental results

In this section we report some experimental results. The experiments were conducted at room temperature. The samples were built of aluminum, niobium, and nickel ultrathin films deposited on various wafers, such as, fused glass, α-quartz and amorphous silicon substrates. The samples were prepared with the sputtering technique assisted by external magnetic field at radio frequency [10]. The thickness of the ultrathin metallic films ranges from 5 to little more than 100 Å. The accuracy of the thickness was estimated to be better than 5%. We have measured infrared optical reflectance as a function of the thickness of the ultrathin films. The incident light was p-polarized with near-normal incident angle. Specifically, the light source was a tunable CO_2 waveguide laser which was linearly p-polarized and tuned at $\lambda = 9.2\mu m$ with an output power of $632mW$. The incident angle was near-normal (7° to the normal $\theta = 7^o$) so as to separate the incident and reflected beams, and the reflectance was lock-in measured as a function of the film thickness. A more detailed description of the experimental setup and the experiments were previously published [11].

In the following three figures, Figs. 1, 2, and 3, curves of measured reflectance as a function of thickness are presented. Three metals, aluminum, niobium, and nickel were tested and the results are included in different figures, Fig. 1 for aluminum, Fig. 2 for niobium, and Fig. 3 for nickel. In each figure, two different curves are presented. The difference between the two curves is the substrate. Three different wafers were used in the experiments, glass, quartz, and silicon.

As already mentioned, the emphasis of the present work is to identify possible quantum size effects. It is understood as follows: If the quantum effects exist, or more precisely, the effects due to quantum confinements are able to manifest significantly enough to stand identifiable from other well-know classis noise sources, such as thermal fluctuation, random scattering, surface roughness, the signs must be tiny and with its unique characteristics, i.e. finger prints.

Before seeking closely for the quantum size effects, one is to study the general tendency of the thickness dependence of ultrathin metallic films. Most of the classic characteristics of the dependence can be understood with well-known knowledge in textbooks on thin metallic films. Some other complications may arise from various sources, such as random scattering and surface plasmons. These are classic effects that may be sensitive to the surface and substrate status and are usually able to bring about smooth changes to the thickness dependence. Previously, we studied the curves for aluminum thin films and

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