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Fourier transform infrared analysis of long-range surface polaritons excited by the end-fire method

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Abstract

The cut-off effect in slightly asymmetric systems prevents the wider practical usage of long-range surface polaritons (LRSP). Due to asymmetry caused by the presence of even a few nanometers wide air gap, the LRSP mode disappears. We propose a solution to this problem by using an effective medium approach. The basic idea is to compensate for the air gap induced asymmetry by introducing an additional layer into the system. That allows us to shift the cut-off to values sufficient for experimental observation of LRSP. Fourier transform infrared spectra of end-fire coupled LRSP in the GaAs/Au/GaAs multilayer system were measured in the (800–3000) cm^{-1} range. Various geometric and polarization sensitive effects of the infrared system capable of influencing the LRSP coupling are discussed. A pronounced increase in the p-polarized light transmission at long-wavelengths is explained as an increase in the propagation distance L of LRSP. A fairly good fit between experimental and calculated curves was achieved at long wavelengths.

Keywords: Surface plasmons; FTIR spectroscopy; Gallium arsenide; Gold

1. Introduction

When a nanometer thickness metal film with complex permittivity $\varepsilon_2=\varepsilon_{r2}+\varepsilon_{i2}$ is located symmetrically between two identical dielectrics (ε_1 and ε_3), the dispersion curve for surface polaritons splits into two branches as a result of interference. The low-wavenumber asymmetric k^- mode suffers low attenuation and for that reason is called the long-range surface polaritons (LRSP). Even in the visible region, the propagation distance L of LRSP can reach a few millimeters. In contrast to the conventional surface polaritons at a single interface, the electric field vector of the k^- mode is always oriented normally to the interface. LRSP are of special interest for various nonlinear and sensoric applications because long interaction lengths and strong polariton enhanced fields can be obtained. These applications have stimulated many spectroscopic investigations of LRSP. However, k^- modes excited by thermal infrared sources have not been reported.

A detailed analysis of LRSP modes in both symmetric and slightly asymmetric systems can be found in [1]. It has been shown [2] that if a thin silver film is embedded in a slightly asymmetric system L increases by several magnitudes, before its cut-off. In order to obtain long propagation distances the LRSP wavevector has to be very close to the cut-off value. Under a slight asymmetry, when $\varepsilon_1-\varepsilon_3=\Delta\ll\varepsilon_1,\varepsilon_3$, the cut-off thickness d_{cut} can be expressed as [3]

$$d_{cut}=\frac{1}{k_0(\varepsilon_1-\Delta)}\left[\frac{\Delta\left(\varepsilon_{r2}^2+\varepsilon_{i2}^2\right)}{\left(\varepsilon_1-\varepsilon_{r2}\right)^2+\varepsilon_{r2}^2}\right]^{1/2}. \quad (1)$$

The maximum value of $\varepsilon_1-\varepsilon_3$, which will give a LRSP mode, is for $\varepsilon_1>\varepsilon_3$,

$$\Delta_{max}=\frac{4\left[\frac{\pi d}{\lambda_0}\right]^2\left[\left(\varepsilon_1-\varepsilon_{r2}\right)^2+\varepsilon_{r2}^2\right]\varepsilon_1^2}{\varepsilon_{r2}^2+\varepsilon_{i2}^2+8\left[\frac{\pi d}{\lambda_0}\right]^2\left[\left(\varepsilon_1-\varepsilon_{r2}\right)^2+\varepsilon_{r2}^2\right]\varepsilon_1}. \quad (2)$$

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It follows from (2) that Δ_{max} is approximately proportional to $1/\lambda_0^2$. Thus, the longer wavelength, the more critical is the matching condition between ε_1 and ε_3. For example, if a system consists of a 20 nm Ag film surrounded by identical dielectrics ($\varepsilon_{1,3}=4.0$) there is a cut-off in the propagation distance of the LRSP mode ($\lambda=10.6$ μm) even in the presence of a 5.2 nm air gap [4]. This corresponds to the substrate flatness $\lambda/100$ at optical wavelengths. The magnitude of d_{cut} sharply depends on the refractive index of the adjacent dielectric media. For example, for a gold film between two plates of GaAs ($\varepsilon_{1,3}=10.89$), the critical air gap equals only 1.8 nm and is practically independent of λ.

In practice, a near-symmetric system can be realized by mechanically pressing a second identical plate (superstrate) against the metal film evaporated on the substrate. This has been successfully realized in several works at the visible and near-infrared wavelengths [5–8]. A large variety of index matching fluids existing for this region allows easy coupling between incident light and LRSP. But this is not the case in the infrared region because of very small values of cut-off thickness and the absence of the high refraction index fluids. To our knowledge, only $n<2.3$ fluids are available nowadays. The refractive indexes of most A^2B^6, A^3B^5 semiconductors, also Si, Ge, possess higher values.

In this work we present first experimental observation of infrared LRSP using Fourier Transform Infrared (FTIR) technique. It will be demonstrated that the cut-off effect caused by the existence of an air gap between metal and dielectric can be eliminated by introducing an additional compensating layer. FTIR spectroscopic experiments in a wide spectral range demonstrate the validity of this effective medium approximation model.

2. Experimental details

2.1. Samples

We have studied experimentally the multilayer system GaAs/Au/air/Ge/GaAs. Two gold films of thicknesses 15 and 20 nm, and one 668 nm germanium film were produced by thermal evaporation on separate 0.5 mm GaAs substrates. The film thicknesses were controlled in situ by the quartz resonator method. Two different samples of 6 mm and 18 mm lengths were cut from the same piece of the substrate. After that two plates were pressed together. In reality, some air gap between Au and Ge films occurs due to the finite flatness of the substrate. An interferometric evaluation indicated better than 100 nm flatness of the GaAs substrate. This procedure was accomplished by pressing the test plate made of high flatness glass to the GaAs plate. A good quality sharp edge of the sample is required for the effective polariton excitation. Quality of this edge was visually controlled by the optical microscope.

2.2. Optical setup

As usual, LRSP are excited using the ATR prism coupler. To our knowledge, all previous experimental works on LRSP were performed using laser sources. The reason for that is the complicated matching of the incident light wavevector with that of LRSP. Even in the visible region the halfwidth of the LRSP induced reflectance minimum occurs in the range of a few angular minutes. In the infrared one it becomes even less than the laser beam divergence. Moreover, due to the surface wave dispersion, only a narrow frequency range can be coupled using a prism method. Under such circumstances, experimental observation of LRSP by a prism method becomes very complicated, and the end-fire method is more preferable [8]. It has been shown numerically that this method leads to an efficient broadband coupling that is surprisingly insensitive to the alignment conditions [8]. Matching of wavevectors between incident light and LRSP is fulfilled automatically. These peculiarities of excitation are essential in case of non-laser spectrometric measurements. Due to the described reasons we chose an end-fire method in our experiments.

Optical sketch of a LRSP accessory is presented in Fig. 1. A precise mechanical x–y–z table for the sample micro-alignment was combined with a Perkin Elmer 1760 series FTIR spectrometer, operating in the external mode. Spectral region ($2000>\nu>750$)cm^{-1} was limited by the transparency region of GaAs at low frequencies and by the lower propagation distances of LRSP at high frequencies. Polarization of the incident beam was selected by the metal grid polarizers made of 0.4 μm period Al wires on the KRS5 substrate (IGP225 model, Cambridge Science, Co.). Measurements of the transmission ratio at two orthogonal polarizations allowed us to escape distortion of the LRSP spectra caused by the direct transmitted beam. We used a $f=2$ cm antireflection coated KRS5 lens L_1 for focusing the incident light onto the end face of a sample. The metal slit D in front of the sample was placed for the purpose to limit the beam size. The magnitude of a slit should be compatible with z-distribution of the polariton field and it was determined by numerical calculations. The field decay is strongly dependent on the gold film thickness and sharply increases when d decreases. As usual, a slit was 0.3 mm width. The sample together with a slit had possibility to be removed in case of the reference signal I_{ref} measurements.

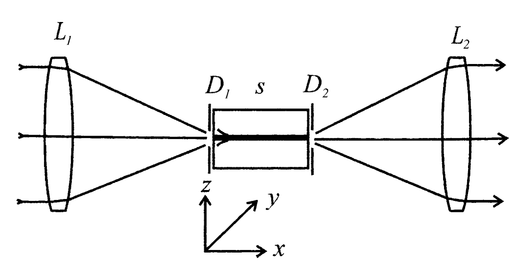

Fig. 1. Sample compartment of the FTIR spectrometer and optical setup for the LRSP excitation.