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Origin of the low frequency radiation emitted by radiative polaritons excited by infrared radiation in planar La_2O_3 films

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Abstract

Upon excitation in thin oxide films by infrared radiation, radiative polaritons are formed with complex angular frequency ω, according to the theory of Kliewer and Fuchs (1966). We show that radiative polaritons leak radiation with frequency ω_i to the space surrounding the oxide film. The frequency ω_i is the imaginary part of ω. The effects of the presence of the radiation leaked out at frequency ω_i are observed experimentally and numerically in infrared spectra of La_2O_3 films on silicon upon the excitation by infrared radiation of the 0TH type radiative polariton. The frequency ω_i is found in the microwave to far infrared region, and depends on the oxide film chemistry and thickness. The presented results might aid in the interpretation of fine structures in infrared and, possibly, optical spectra, and suggest the study of other similar potential sources of electromagnetic radiation in different physical scenarios.

1. **Introduction**.

Nature has developed various mechanisms to produce radiation, such as blackbody radiation [1, 2], inter-band electron transitions between energy levels in atoms or molecules [3-5], the acceleration of electrons or charged particles [6-11], radiative nuclear reactions [12], and Cherenkov radiation [13, 14]. Recently, radiation was found to be originated by quasi-particles called polaritons. Specifically, photoluminescence [15] and laser radiation [16] in the near infrared (NIR) and visible regions were observed at low temperature (~ 10 K) in periodically patterned semiconducting samples and micro-cavities, where polaritons can propagate. Here we describe an additional route nature follows to produce radiation from polaritons: the outburst of far IR (FIR) or microwave radiation by radiative polaritons (RPs) at room temperature in simple planar dielectric films, where polaritons cannot propagate. Indeed, RPs [17] and radiative plasmons [18, 19] are unable to propagate in planar dielectric layers because of the lack of periodic structures acting as wave-guides and enabling constructive interference [20]. The RPs have a group velocity faster than the speed of light in vacuum c [17-19], but their inability to propagate suggests that they do not behave as superluminal wave pockets [21, 22]. Therefore, this work describes the energy-release mechanism in RPs with group velocity faster than c upon generation in planar dielectric layers illuminated by IR radiation. Radiative polaritons are widely used to aid in the interpretation of IR spectra [23-26], nevertheless the origin of the radiation generated during their excitation by IR radiation in thin oxide films or slabs has so far not been investigated.

2. **The hypothesis on the origin of the radiation emitted by radiative polaritons**.

We consider RPs excited in thin planar oxide films [23] by IR radiation [17]. The dispersion relation in Fig. 1, where ω and k_x are real quantities, is divided into two regions. The surface phonon-polaritons (SPPs) [20, 27] appear to the right-hand side of the light-line $\frac{\omega}{k_x} = c$ in the $\omega - k_x$ plane, where ω is the angular frequency, and k_x is a component of the wave-vector $\mathbf{k}$ parallel to the oxide film surface [17, 20]. The SPPs are generated in dielectric layers with periodic structures, where they can propagate [28]. On the other hand, the RPs appear to the left-hand side of the light-line in Fig. 1. The left-hand side of the light-line is the long-wavelength (λ) section of the dispersion relation plane, where $k_x = \frac{2\pi}{\lambda} \to 0$ [17, 20]. Moreover, the prominent 0TH type-RP of interest here appears next to the longitudinal optical (LO) phonon frequency ω_{LO} [17, 24]. At this frequency, the oxide dielectric function $\varepsilon(\nu)$, where ν is a real frequency, reaches a minimum. In this condition, if the polariton would propagate, its group (v_g) and phase (v_{ph}) velocities would coincide, such that $v_g = v_{ph} = \frac{\omega}{k_x} > c$ [17]. However, the dielectric layers supporting RPs are planar. They do not have periodic structures acting as wave-guides and enabling constructive interference, which would promote RPs propagation [15, 16, 28]. Therefore, RPs dissipate [29] their potential energy and conserve their momentum, $|\mathbf{p}| = \frac{\hbar\omega}{c}$, by acquiring a complex angular frequency $\omega = \omega_r + I\omega_i$ and wave-vector $k_x = k_{xr} + Ik_{xi}$ [17, 18, 30, 31]. In these expressions, I is the imaginary unit and the subscripts r and i refer to real and imaginary parts, respectively. Radiative polaritons absorb IR radiation at ω_r, as observed in thin planar dielectric films and slabs [23-26]. In this research, we

address the energy-release mechanism enfolded in the existence of the imaginary angular frequency ω_i of RPs described as plane waves with complex angular frequency ω [17].

The current literature associates imaginary angular frequencies with phase instabilities, for example in phase transitions [32], and to waves moving away from their source, for example in quasi-normal modes in gravitational waves [33]. For RPs, the physical implications of the existence of the imaginary frequency ω_i are rooted in their polarization vector **P**, which has a plane wave type of space/time dependence [17, 20]. Therefore $\mathbf{P} \propto e^{i(k_x x - \omega t)}$, and, since for RPs ω and k_x are complex [17], it occurs that:

$$\mathbf{P}(x,t) \propto e^{i(k_{xr}x-\omega_r t+ik_{xi}x-i\omega_i t)} = e^{i(k_{xr}x-\omega_r t)}e^{-(k_{xi}x-\omega_i t)}. \tag{1}$$

Physically, ω_r and ω_i can be determined from the center and half-width of the peaks or dips generated by RPs in IR spectra of oxide films or slabs [17, 24]. The ω_r values are found near the frequencies of transverse optical (TO) or LO phonons [17, 24]. Since for RPs $\left|\frac{\omega}{k_x}\right| > c$, it occurs that $|k_x| = \frac{2\pi}{\lambda} \to 0$, and that k_{xr} and k_{xi} are small. Thus, it can be assumed that the imaginary parts ω_i and k_{xi} satisfy $\frac{\omega_i}{k_{xi}} > c$ and $k_{xi}x < \omega_i t$. Therefore, the term $e^{-(k_{xi}x-\omega_i t)}$ in Eq. (1) has a positive exponent which increases with t and distance. Due to the similarity in behavior predicted for other systems with imaginary frequency [33], we hypothesize that the portion of the polarization vector $\mathbf{P}(x,t) \propto e^{-(k_{xi}x-\omega_i t)}$ corresponds to radiation leaked at an angular frequency around ω_i to the space surrounding the oxide layer where the RPs are excited by IR

radiation. Since k_x is complex, the direction of propagation of the radiation leaked to the surrounding space is uncertain [17].

3. Experimental determination and characterization of the frequency ω_i of the radiation emitted by radiative polaritons.

To experimentally determine the frequency range of the ω_i values, we examine the IR spectra versus the IR radiation angle of incidence θ_0 [17] for the 0TH type-RP excited in thin oxide films, 250 nm thick or less, on metallic and semiconducting substrates. For this work, thin planar amorphous Al_2O_3 films were deposited by atomic layer deposition (ALD) [34] as described in Ref. 23. The γ-Al_2O_3 crystalline structure [35] was obtained by heat treatment for one hour at 600°C of the as-grown film. The reflectance spectra of the Al_2O_3 film on Al foil and Si(100) were acquired in the middle IR (MIR) region (650-7500 cm^{-1}) using a N_2 purged Bruker Vertex 70 Fourier transform IR spectrometer (FTIR). The broadband IR radiation is generated by a globar (Q301) source. The samples were placed on the variable angle (30-80°) reflection accessory Veemax II by Pike Technologies, with a gold mirror on the back, in contact with the substrate. In the reflectance spectra obtained with transverse magnetically polarized IR radiation for the 250 nm thick γ-Al_2O_3 films on Al foil and Si(100), the 0TH type-RP corresponds to the dip at ω_r between 960 and 975 cm^{-1} [23, 36]. The polarization is achieved using a ZnSe polarizer. The measurements were carried out with a 2 cm^{-1} resolution.

Data collected for a 250 nm thick γ-Al_2O_3 film on Al foil [23] and Si(100) in the 30 to 70° θ_0 range indicate the increase of the experimental ω_i values from approximately 30 to 48 cm^{-1} for the 0TH type-RP. Beyond θ_0=70°, the uncertainty of the ω_i values increases because

of the small percentage of IR beam cross section effectively exploited in the measurements at grazing angles. In IR spectra, all oxides exhibit dips with similar widths [23-26, 36]. Generally, the ω_i values in thin oxide films are found in the microwave to low FIR frequency range (10-450 cm^{-1}), and are only slightly substrate-dependent.

To discern whether the ω_i values depend on oxide film chemistry, the results for the 250 nm thick γ-Al_2O_3 films are compared with same-thickness films of anatase (a)TiO_2 on Al foil with $\omega_r \cong 850$ cm^{-1} [37], and hexagonal (h)La_2O_3 on Si(100) with ω_r = 546 cm^{-1} [38]. The a-TiO_2 film on Al exhibits an ω_i of ~ 40 cm^{-1} at θ_0=75°, while the h-La_2O_3 film on Si(100) shows an ω_i of ~ 50 cm^{-1} at θ_0=70°. Thus, a slight dependence of ω_i on oxide film chemistry exists. In addition, we observe a dependence on film thickness of the ω_i values. The data indicate that, at θ_0=65°, the ω_i values for 250 and 50 nm thick γ-Al_2O_3 films on Al foil are ~ 37.5 and 34.0 cm^{-1}, respectively. There exists an even larger difference for a-TiO_2 films on Al foil. A 50 nm thick film with ω_r = 863 cm^{-1} exhibits an ω_i value of 18.8 cm^{-1} at θ_0=65°, which is approximately one half of the ω_i for the 250 nm thick film. Further evidence of the slight oxide film thickness dependence of ω_i is offered by the 40 and 20 nm thick Lu_2O_3 films on Si(100) and CoSi [39] indicating at θ_0=65° an ω_i of ~ 12 and 18 cm^{-1}, respectively. The slight oxide chemistry and thickness dependence suggest that the ω_i values are correlated to the amount of RPs excited in the oxide film and to the atomic weight a_m of the metal ion in the oxide such that, approximately, $\omega_i \propto \sqrt{a_m}$, as shown in Fig. 2 for 250 nm thick films. This trend suggests that RPs behave differently than harmonic oscillators, and that their production resembles a "burst" that channels out energy. Similar phenomena, such as electromagnetic emissions from strong

turbulences [40] or electron expulsion [41] accompany plasma waves when their phase velocities approach the relativistic regime.

4. Detection of the radiation emitted by the 0TH type radiative polariton in La_2O_3 films on silicon.

The experimental detection of the radiation leaked to the surrounding space by the excitation of RPs is challenging, (1) because of the uncertainty of the direction of propagation of the radiation leaked to the surrounding space [17], and (2) because it is difficult to find an oxide such that its 0TH type-RP exhibits both the ω_i and ω_r frequencies in the FIR region, where they can be contemporarily detected. Problem (1) is solved by performing the measurements keeping the detector in a fixed position, and varying the IR radiation incidence angle θ_0 on the oxide film surface. The expected signature of the radiation leaked to the surrounding space is a dip or a peak at the ω_i frequency in the transmittance or reflectance spectra. The dip or peak witness the fact that the radiation leaked to the surrounding space by the excitation of RPs is ruled neither by Snell's law nor by the law of reflection. Problem (2) is solved choosing La^{+3} as the metal ion, which is sufficiently heavy to generate oxides characterized by a prominent 0TH type-RP with the ω_i and ω_r frequencies in the FIR region. Therefore, two 250 nm thick La_2O_3 films on Si(100) are selected for the investigation, one with hexagonal (h), and the other with cubic (c) phase. The h-La_2O_3 film on Si(100) was grown by ALD, as described in Ref. 38, extracted from the reactor, and immediately annealed in vacuum at 600°C for 60 s. The c-La_2O_3 film on Si(100) was similarly grown by ALD, but was capped with a layer of ALD Al_2O_3 about 5 nm thick before the extraction from the reactor [42]. Afterwards, the capped film was immediately annealed in vacuum at 600°C for 60 s. Transmittance FTIR spectra were then collected for these

samples. The schematic of the experimental set-up is shown in Fig. 3. The transmittance FTIR spectra in Fig. 4(a) were measured in the FIR region using non-polarized IR radiation, as described in Ref. 38, in a 1 mbar sample compartment at θ_0=0° and 70°. The ω_i values for the 0TH type-RPs are ~ 50 cm^{-1} at θ_0=70° for both h- and c-La_2O_3 films on Si(100). The spectra at θ_0=70° for the 250 nm thick h- and c-La_2O_3 films on Si(100) exhibit a dip at 55.5 cm^{-1}, clearly shown in Fig. 4(b). The dip is slightly attenuated by the Al_2O_3 capping layer on c-La_2O_3. The spectra at θ_0=0° for the same films exhibit no dip. Within the errors, the location of the dip in spectra collected at θ_0=70° in Fig. 4(b) correlates with the ω_i values for the examined films (~ 50 cm^{-1}, as reported in Sec. 3). Thus, we consider the dips in the transmittance FTIR spectra as the signature of the radiation leaked to the surrounding space following the excitation by IR radiation of the 0TH type-RP in the 250 nm thick h- and c-La_2O_3 films on Si(100). The dip was simulated in Figs. 4(a) and (b) by adding to the expression of transmittance from Fresnel's equations [43], a term proportional to $-\frac{\omega_i}{\nu-\omega_i}$, where ν is the real frequency on the abscissa of the IR spectra. A detailed description of the simulation of the transmittance spectra and corresponding fine structures in the FIR region of the h-La_2O_3 films on Si(100) is given in the Supplementary Material [44].

5. **Summary and significance**.

In summary, we have described a source of electromagnetic radiation which accompanies the formation of an object, e.g. a radiative polariton in a thin planar oxide film, characterized by the inability to propagate in the layer supporting its excitation. Since radiative polaritons have long wavelength λ and large frequency ω such that their group velocity, $v_g = \omega\lambda$, is faster than

c, the speed of light in vacuum, upon excitation they dissipate their potential energy into radiation by acquiring a complex frequency. Bursts of low frequency radiation originate from the dissipation process accompanying the excitation of radiative polaritons. The imaginary part of the complex frequency of the radiative polaritons determines the magnitude of the frequency of the radiation given off.

The presented results are relevant (i) for stimulating the search for radiation leaked to the surrounding space by radiative plasmons in metallic layers, which have similarities with the radiative polaritons, and (ii) for explaining the fine structures in infrared and, possibly, optical spectra. This work could suggest the study of other similar potential sources of electromagnetic radiation in different physical scenarios, such as a beam of high frequency photons colliding with a beam of low frequency–large wavelength radio-waves, or a beam of high frequency photons in space colliding, for example, with gravitational waves with low frequency and large wavelength.

Acknowledgments

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44. Supplementary Material.

Figure Captions.

Fig. 1. Polariton dispersion relation showing the real angular frequency ω versus the real wave-vector k_x normalized with respect to the values for TO phonons [17]. The index x indicates a direction parallel to the oxide layer surface. The RPs and SPPs appear to the left- and right-hand side, respectively, of the light-line in vacuum $\frac{\omega}{k_x} = c$ in the $\omega - k_x$ plane, [17]. As described in Ref. 17, RPs appear in regions labeled R_1, R_2, and $R_1^{'}$, SPPs appear in regions labeled L_1, L_2, and $L_1^{'}$, and no features exist in the regions labeled N. The dielectric constants $\varepsilon(0)$ and $\varepsilon(\inf)$ are the values of $\varepsilon(\omega)$ for zero and infinite frequency.

Fig. 2. Approximate dependence of ω_i on the atomic weight a_m of the metal ion in the oxide such that $\omega_i = \left(3.3\sqrt{a_m} + 25\right)$ cm^{-1} evaluated for 250 nm thick oxide films. The experimental ω_i values were measured with an uncertainty of ~5 cm^{-1}.

Fig. 3. Schematic of the experimental set-up for the IR transmittance measurements at an angle of incidence θ_0 with respect to the normal **n** to the oxide film surface and the (x-y) plane. The (x-y) plane is the plane of incidence of the IR radiation on the thin oxide film surface. The incident and transmitted IR radiation is indicated by the wave-vectors $\mathbf{k}_{INC}$ and $\mathbf{k}_{REFR}$, respectively. The radiation leaked to the surrounding space following the excitation by IR radiation of the 0TH type-RP in a La_2O_3 film on a substrate is represented by the waves with wave-vector $\mathbf{k}_S$.

Fig. 4. (a) Experimental (ex.) and simulated (sim.) transmittance FTIR spectra measured at θ_0 =0° and 70° in the FIR region using non-polarized IR radiation. The experimental spectra were

obtained as described in Ref. 38 for 250 nm thick h-La_2O_3 and c-La_2O_3 films on Si(100). The simulated spectra were obtained as described in the Supplementary Material [44] for a 250 nm thick h-La_2O_3 film on Si. For clarity, the spectra are given an arbitrary offset. (b) Zoom in the 50-150 cm^{-1} range of the spectra in panel (a). The symbol h- indicates the h-La_2O_3 film on Si(100), whereas c- indicates the c-La_2O_3 film on Si(100). In panels (a) and (b) and in the spectra measured at θ_0=70°, the arrow indicates the dip located at 55.5 cm^{-1} which correlates, within the errors, to the ω_i values for the examined La_2O_3 films.

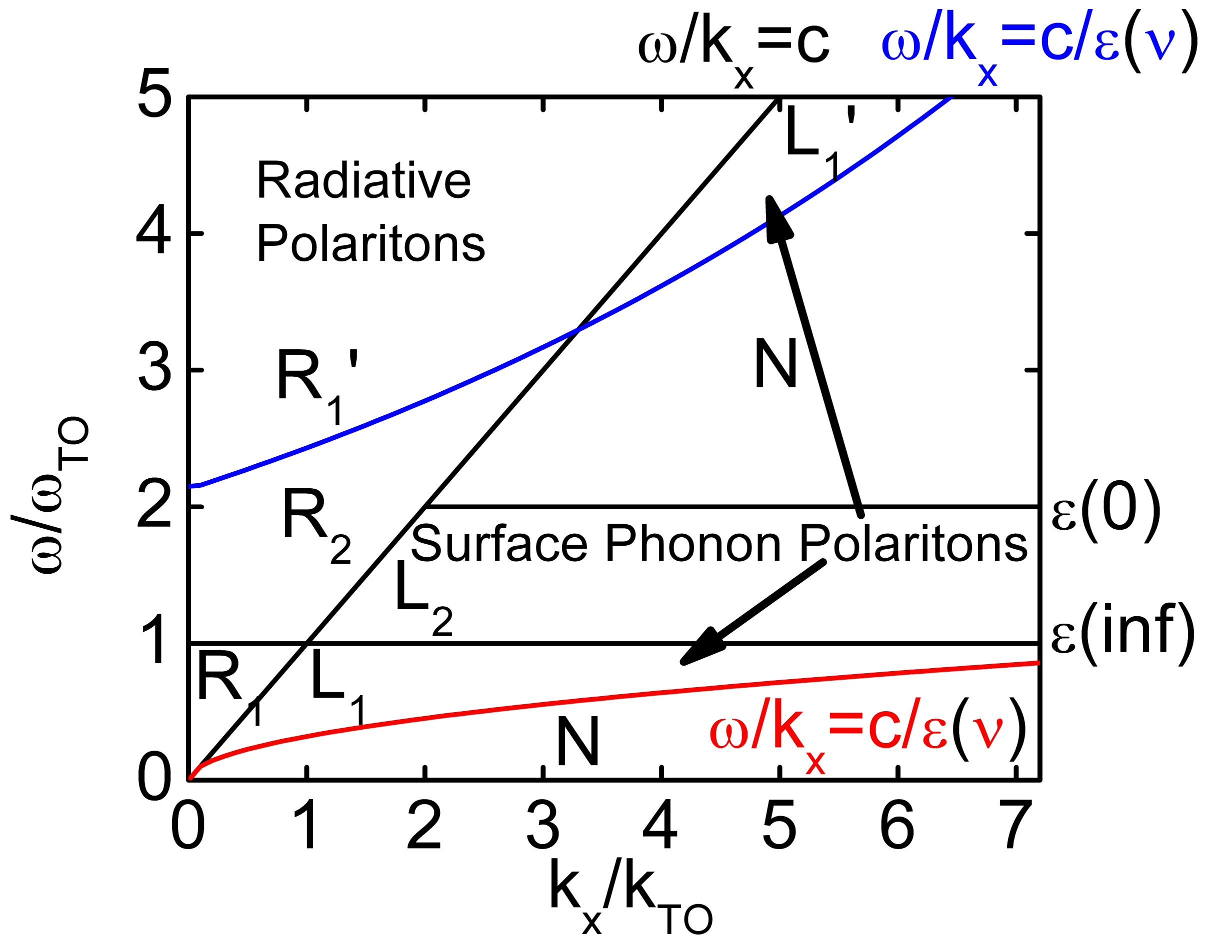

Fig. 1

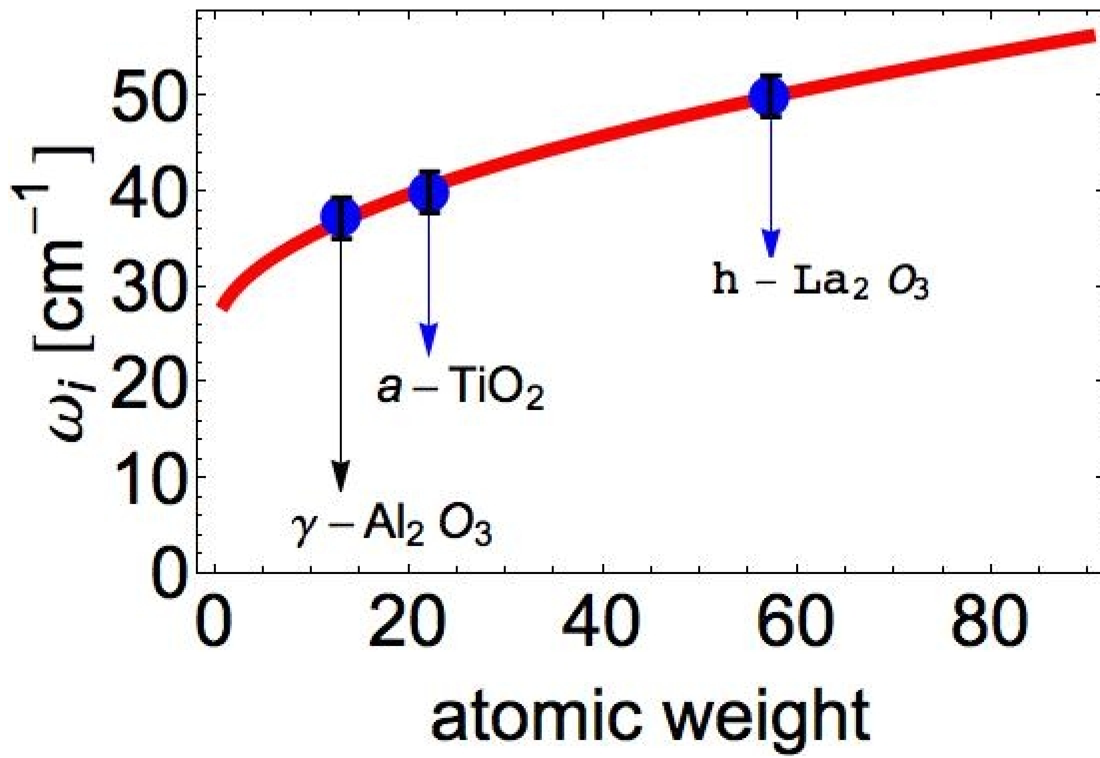

Fig. 2

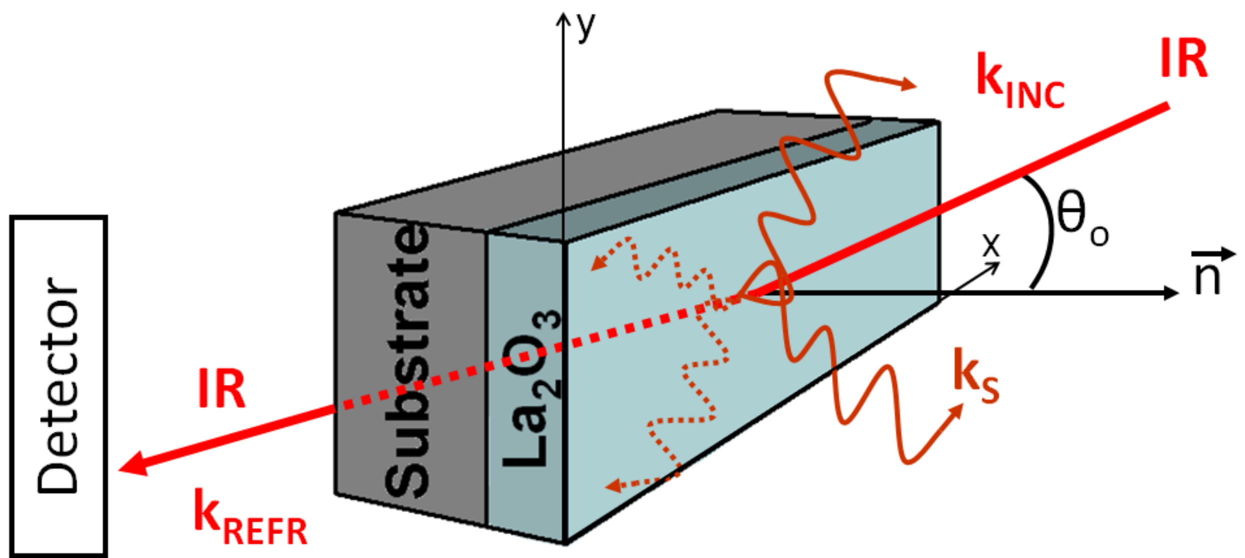

Fig. 3

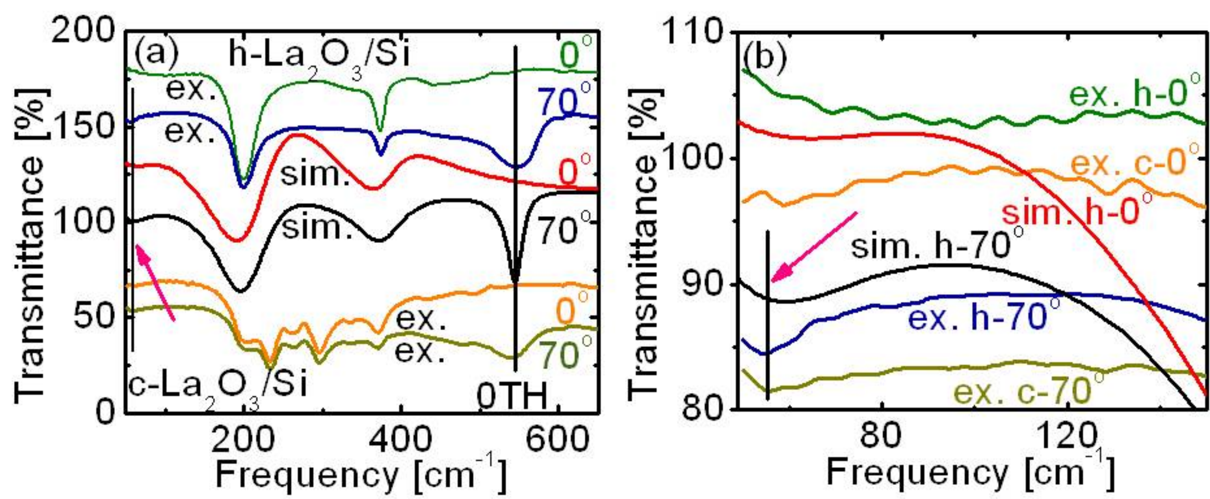

Fig. 4