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Near-field Thermal emission of sapphire at high-T: precisely locating the T-dependent polariton resonances in view of energy conversion

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Abstract: *In the near-field of a hot ($T \leq 1100$ K) window, we use atom spectroscopy as an ultra-narrow spectral filter to probe the T-dependent surface polariton resonance of sapphire ($\sim 12.2 \mu\text{m}$). The results agree only partly with predictions derived from broadband far-field emissivity. Sapphire birefringence makes predictions difficult, but adds to versatility. Near-field thermal emission could be converted into a molecular excitation, opening a path to electrical conversion.*

Keywords Near-field, Surface polariton, Casimir-Polder interaction birefringence

The central topic of this presentation concerns information on the spectral location and features of surface polaritons (sapphire in our case) which are at the origin of the well-known enhancement of thermal exchanges in the near-field [1]. A surface polariton is the evanescent electromagnetic mode which describes the thermal field at an interface between material and vacuum in the extreme near-field, *i.e.* distances much shorter than the wavelength. Considering the interface between vacuum and a (homogeneous) plate, described by a (relative) dielectric permittivity, the key information for the relevant surface mode is contained in the complex "(planar) surface response".

$$S(\omega) = [\varepsilon(\omega) - 1] / [\varepsilon(\omega) + 1] \quad (1)$$

The dissipative part of this surface response, governed by $\Im m S$, allows a selective transfer of energy to passing-by particles. This transfer occurs if the particle can resonantly absorb the polariton mode. This scheme may convert thermal radiation, from a classical source for broadband lamps for far-field, into a selective pumping lamp in the near-field. With such an optical pumping induced by near-field thermal radiation, conversion into electrical energy can be the next step, through the thermally induced chemical or optical excitation.

Among standard materials easily operated at high-temperatures, SiC, used for the first demonstrations of the near-field properties of the thermal radiation, exhibits a (near-field) narrow peak around $10.5 \mu\text{m}$ (28.5 THz, quality factor ≤ 100). Sapphire also accepts very high temperatures, with its (near-field) peak around $12.2 \mu\text{m}$ (~ 24.5 THz) [2]. Note that the precise features of the surface thermal emission vary with temperatures, because the phonons, and hence the surface polariton of the materials, evolve phenomenologically with temperature [3,4], aside from the thermal statistics governing the overall thermal field.

The evaluation of the surface response $S(\omega)$ is usually indirect, as derived from the knowledge of the permittivity $\varepsilon(\omega)$ [becoming $\varepsilon(\omega, T)$ if the temperature-dependence is available]. Moreover, the permittivity $\varepsilon(\omega)$ is most often evaluated from far-field emittance (or reflectance [3]), through a fitting procedure based upon a more or less elaborate model. Alternately, direct measurements for $\varepsilon(\omega)$ are sometimes feasible (*e.g.* with ellipsometry), but for a limited range of frequency (usually requiring partial transparency), and temperature.

This indirect method was used for a sapphire plate in experiments performed at CEMHTI, where equipment specifically devoted to high-temperature emittance is available [5], and with an elaborate multi-parameter model [3]. A principle difficulty in the evaluation of the

surface response is that its resonances are located in a range where the permittivity spectrum is very unspecific, and that the salient features of the permittivity itself do not exhibit any resemblance with the major features of the original reflectance or emittance.

On a very similar sapphire plate (same crystal bowl, same super-polishing and annealing), an original Atomic Physics method was used at LPL as a diagnostics. It essentially involves an ultra-narrow atomic filtering of the near-field thermal emission.

The team at LPL is familiar with the probing of long-range atom-surface interaction in the (van der Waals) dipole-dipole interaction limit of the "Casimir-Polder" interaction [6]. Through a detailed analysis of a spectral lineshape -through selective reflection spectroscopy [2,6], ensuring the probing of the vicinity of the surface-, we can evaluate, for a given atomic level, the strength of the potential $-C_3z^{-3}$ exerted onto the atom by the surface. In particular, C_3 had been shown to depend on temperature owing to thermal statistics [2], and on the specific resonant coupling between a (virtual) atomic emission and a surface polariton mode [7]. Here, the excited Cs level $7P_{1/2}$ is coupled in absorption a 24.687 THz transition to $Cs(6D_{3/2})$, and the measured temperature evolution of $C_3(T)$ is essentially dependent on $\Re\epsilon(\omega)$ [for $\omega=24.687(2\pi)$.THz]. Thanks to our lineshape analysis, we extract a rapid temperature evolution of $C_3(T)$. Its growth slows down at high-temperature, because of a broadening of the sapphire polariton. As a first result (fig.1), LPL measurements appear more compatible with CEMHTI measurements, and other T-dependent evaluations, than with predictions derived from the standard thermal statistics, and considering for sapphire only the room-temperature properties.

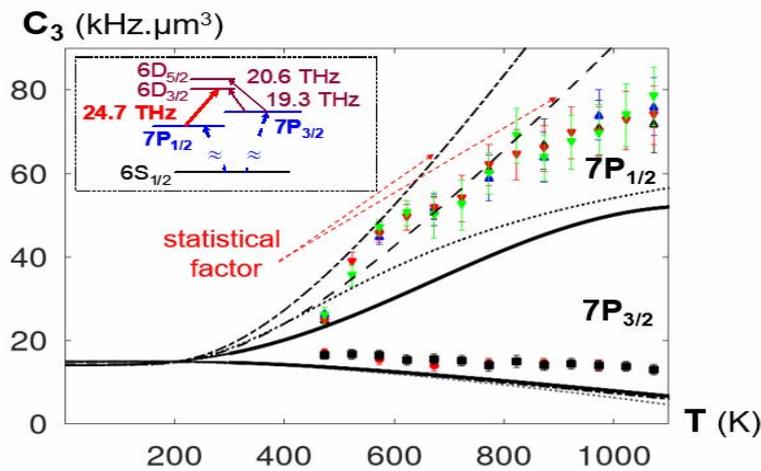


Figure 1: $C_3(T)$ as extracted from atomic spectroscopy (points) on the Cs transitions $6S_{1/2}$ - $7P_{1/2}$ and $6S_{1/2}$ - $7P_{3/2}$ (see inset), along with various predictions (lines) derived assuming various models for the sapphire permittivity $\epsilon(\omega)$. The dashed [8] and dashed-dotted [9] lines use sapphire permittivity as determined solely at room temperature, with $C_3(T)$ only governed by a thermal statistics factor; the dotted line [4] and the thick line -our measurements at CEMHTI- involve a temperature-dependence for sapphire permittivity.

Similar spectroscopic measurements were performed on the other fine-structure component $Cs(7P_{3/2})$, whose dominant couplings to $Cs(6D_{3/2})$ and $Cs(6D_{5/2})$ (respectively at 19.3 THz and 20.6 THz) fall only on the wings of the sapphire polariton resonance; This makes $C_3(T)$ predictions rather insensitive to the detailed modeling of the sapphire polariton. Aside from the atom-surface potential $-C_3z^{-3}$, the resonant atom-surface polariton coupling can induce a *real* absorption of the surface polariton into an atomic excitation towards $Cs(6D_{3/2})$; no equivalent exists for $Cs(7P_{3/2})$. The transfer rate, naturally depending on temperature, is governed by a $\Im\epsilon(\omega).z^{-3}$ dependence [with $\omega=24.687(2\pi)$.THz]. This effect induces a change in the spectroscopic lineshape, which is nearly equivalent to a decrease of the reflected amplitude -by a factor ~ 2 for our temperature range. Because of the linearity of our spectroscopic method, the amplitude comparison, between a study for $Cs(7P_{1/2})$ and for

Cs(7P_{3/2}) is significant. The expected decrease is actually not observed, pointing out a disagreement with the predictions derived from far-field measurements.

This discrepancy for $\Im S(\omega)$ has to be considered in line with the fact that even for a moderately sharp surface resonance $S(\omega)$, the complex response is locally (*i.e.* around resonance) a complex Lorentzian [10]. The acceptable comparison between our two methods for $\Re S$, along with the strong divergence for $\Im S$, suggest that the exact location of the surface polariton is shifted, with respect to predictions from far-field measurements. In particular, the already sharp value for $\Re S(\omega)$ [at $\omega = 24.687 (2\pi)$.THz] should not be considered as found in the "abnormal dispersion" region of the dispersive response .

As a specific difficulty of sapphire, it is birefringent. This imposes to average the surface response of a $c_{//}$ and of a c_{\perp} window. Indeed, for the atomic physics experiment, the window is c_{\perp} -cut (to ensure the cylindrical symmetry of the surface interaction) while field fluctuations have to be integrated over all directions. In the far IR, the optical differences associated to the choice of axis orientation are much more considerable than in the visible range. Moreover, sapphire is known to exhibit several phonon modes (4 or 2, depending on the orientation). This induces uncertainty when evaluating $S_{eff}(\omega)$, as derived from eq.(1) with $\epsilon_{eff} = (\epsilon_o \cdot \epsilon_e)^{1/2}$.

In conclusion, we show that accurate spectral predictions of the surface polariton exhibit various difficulties, possibly of a high-impact to realize an efficient energy transfer in the near-field, as it can be considered with hot SiC for the selective vibrational excitation of molecules such as NH₃ or SF₆ [NB: for our Cs experiment, Cs(7P_{1/2}) is populated only after a prior excitation from the ground state, and the best energy transfer would be a kind of up-conversion [11]]. Accurate evaluation of the surface resonances can be also critical for various near-field systems involving thermal diodes. Finally, thermal exchanges between two different crystalline cuts of a single material may exhibit interesting properties, whose predictions remain delicate.

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