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Polarization pyrometry – an improvement to multi-wavelength optical pyrometry

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Abstract
We describe a new method that improves upon temperature measurement by optical pyrometry. The main uncertainty in the traditional pyrometry technique is the surface emissivity, which is generally unknown and hard to measure. A common approach to deal with this problem is to measure the thermal emission at multiple wavelengths – an approach called multi-wavelength pyrometry. However, this technique can still result in a level of uncertainty in the surface temperature that is unsatisfactory for scientific applications, such as a measurement of equation of state of warm dense matter (WDM). In contrast to the conventional multi-wavelength technique, in the polarization pyrometry approach described herein, p- and s-polarization components of thermal radiation at multiple-angles are used to deduce the temperature. This paper describes the concept and the results of an initial proof-of-principle static experiment with an electrically heated tungsten ribbon. It was found that in same experiment, the accuracy of the polarization pyrometry measurement was substantially greater than that achieved using conventional multi-wavelength pyrometry.

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I. Introduction

A reliable temperature measurement is a key diagnostic in many industrial and scientific applications. Of special interest is the temperature measurement of a sample in a warm dense matter (WDM) state, which is characterized by densities near solid-state, temperatures up to 100,000 K, and pressures up to few Mbar. WDM is relevant to research on plasma physics, astrophysics, geophysics, planetary sciences, and other fields. WDM can be produced with several heating techniques, including lasers, pulsed electric currents, electron, ion and x-ray beam heating, and shock-wave heating.

Pyrometry, determination of surface temperature by analysis of thermally emitted light, is a well-known and commonly used method for temperature measurements of matter in the WDM regime. The pyrometric technique is based on the measurement of thermal radiation $I(\lambda)$, and "comparison" of this $I(\lambda)$ to that of black body (Planck) radiation:

$$I(\lambda) = \varepsilon(n,k,\lambda,\alpha = 0^\circ) \times \frac{C_1}{\lambda^3} \frac{1}{e^{C_2/\lambda T} - 1}.$$ 

**Equation 1:** Intensity of thermal radiation, $I(\lambda)$ at wavelength, $\lambda$ and normal incidence, $\alpha=0$, is a product of a normal spectral emissivity, $\varepsilon$, and black-body radiation. Here n and k are refraction and extinction coefficients of material at wavelength $\lambda$, respectively. $C_1$ and $C_2$ are constants.

The pyrometric method has been used for more than 200 years and has been applied in various applications, ranging from metal casting (~1000 K) to shock compression (~10,000 K) and to intense-laser heated hohlraum targets in inertial fusion research (>10,000,000 K). Despite its wide use, pyrometry has a major difficulty: in order to solve Equation 1 for $T$, one needs to know the emissivity, which is in most cases unknown and must be either measured or approximated.

The emissivity of an ideally clean and smooth surface is a function of the optical constants of the surface material, n and k (the refraction and extinction coefficients). These constants are determined by the AC electrical conductivity, which, in turn, depends on the temperature and density, and hence on the EOS. Exact calculation of the emissivity is possible only in a small region of a phase diagram, where the conductivity is known or reliable models exist. In the high energy density region, such as WDM, not much is known about conductivity, and thus the emissivity must be either measured or approximated. In practice, the commonly used black body approximation, $\varepsilon(\lambda) = 1$ (single-channel pyrometry) is accurate only for certain materials in a narrow temperature and wavelength range. A more advanced technique is multi-wavelength/channel pyrometry, in which radiation is measured simultaneously at multiple wavelengths and a polynomial dependence of $\varepsilon(\lambda)$ on $\lambda$ is assumed (this also includes the case of the "grey body" approximation, in which $\varepsilon(\lambda)$ is constant). The temperature is determined from a fit, in which the polynomial coefficients and the temperature are fitted variables. Fundamentally,
this multi-wavelength approach is flawed. There are N measurements but N+1 unknowns – N unknown emissivities, \( \varepsilon(\lambda) \) at each new wavelength \( \lambda \), and a temperature – resulting in an underdetermined system of equations. In order to reduce the number of unknowns, so that system of equation can be solved “uniquely”, a polynomial (or other) law must be assumed. In a few cases, in which a qualitative emissivity behavior is known a priori – for example when it depends weakly on wavelength, a grey body approximation is suitable, or the emissivity has a smooth polynomial behavior – multi-wavelength pyrometry delivers reasonably reliable measurements\(^9\). However, the emissivity in the WDM regime is not known and can be a complicated function with several extrema or “sharp” features (as, for example, in the emissivity of gold), leading to large, 20%-40%, errors\(^10\) – this is unsatisfactory precision for accurate EOS measurements.

At present, there is no reliable, universal method to directly measure the temperature with better accuracy using the pyrometric approach. This results in several negative implications for WDM experiments. In a typical WDM experiment, the energy, \( E_{\text{dep}} \) is deposited rapidly, on a nanosecond/sub-nanosecond time scale, and is accurately determined. If the temperature cannot be measured with sufficient precision, the specific heat \( C = \frac{dE_{\text{dep}}}{dT} \) is not determined accurately and \( C \) is a key unknown aspect of the equation of state (EOS) of the hot material. At least for certain materials at WDM conditions, phase transitions are expected. Such transitions typically occur at definite temperatures, and the accurate measurement of the transition temperature is obviously a key step for building up the quantitative science of WDM. Finally, almost all of the computer codes used to predict the properties of WDM employ density and temperature as inputs; for this reason it will be much easier to directly compare theory and experiment if one can accurately measure the temperatures attained in any given experiment.

This paper consists of two parts. The first part explains the concept of the polarization pyrometer, discusses its advantage and limitations. The second part summarizes results of an initial test of the technique in low temperature, static experiments.

II. Principle of polarization pyrometry
A conceptual layout of a polarization pyrometer is shown in Figure 1: thermal emission from a sample is collected at several angles, split into two polarization components with polarization optics, filtered spectrally, and focused onto a photodetector.

The emissivity, \( \varepsilon(\lambda) \) of a smooth and clean surface is related by Fresnel formulas to the complex dielectric function\(^4\), \( \xi(\lambda) \). For a non-zero angle \( \alpha \) (to the surface normal), the \( \varepsilon(\lambda) \) is different for the \( p \) and \( s \) components of an electrical vector and the spectral emission intensity of each polarization component can be written as
Equation 2: Spectral radiation density of p- and s-polarization components at angle $\alpha$ and wavelength, $\lambda$; $C_1$ and $C_2$ are the Plank formula constants.

$$I^p_\alpha(\lambda) = \left(1 - \frac{\xi(n,k) \cdot \cos(\alpha) - \sqrt{\xi(n,k) - \sin^2(\alpha)}}{\xi \cdot \cos(\alpha) + \sqrt{\xi + \sin^2(\alpha)}}\right)^2 \times \frac{C_1}{\lambda^\frac{3}{2}} \frac{1}{e^{\frac{C_2}{\lambda}} - 1},$$

$$I^s_\alpha(\lambda) = \left(1 - \frac{\cos(\alpha) - \sqrt{\xi(n,k) - \sin^2(\alpha)}}{\cos(\alpha) + \sqrt{\xi(n,k) - \sin^2(\alpha)}}\right)^2 \times \frac{C_1}{\lambda^\frac{3}{2}} \frac{1}{e^{\frac{C_2}{\lambda}} - 1},$$

$\xi = (n - ik)^2$ – complex dielectric function.

As can be seen clearly in Equation 2, if the absolute intensities of the polarization components, $I^\alpha_{p,s}(\lambda)$ are measured for (at least) two angles (four values in total), there is a unique solution for $T, n, k$. Hence, in addition to temperature, the described concept allows for a direct measurement (without an external light probe) of the optical constants as well as the spectral emissivity.

For improved accuracy, one can always take data at more than two angles and obtain an over-determined system of equations — a favorable condition for measurements with finite accuracy. From the mathematical point of view, polarization pyrometry is a more "powerful" technique than multi-wavelength pyrometry: if measurements are made at more angles, the accuracy progressively increases, while an increase in number of measured wavelengths does not necessarily result in a more accurate value\(^9\).

An equivalent but technically less complex approach is, instead of measuring the absolute intensities $I^\alpha_{p,s}(\lambda)$, measuring the two dimensionless ratios $R_1(\alpha)$ and $R_2(\alpha)$ shown in Equation 3. Combining these ratios with an absolute measurement of the emission at normal incidence, it is similarly possible to uniquely solve the resulting system of equations. An advantage of this approach is that only the $0^\circ$ measurement must be absolute, making the calibration process less elaborate.

$$R_1(\alpha) = \frac{I^p(\alpha)}{I^s(\alpha)}, \quad R_2(\alpha) = \frac{I^p(\alpha) - I^s(\alpha)}{I^p(\alpha) + I^s(\alpha)}.$$  

Equation 3: Dimensionless ratios that can replace the absolute measurements and greatly simplify the calibration of the instrument.

According to Equation 2, the s-component of the thermal emission is always weaker than the p-component. This is also obvious in Figure 2, where the numerically calculated angular dependence of the polarization resolved emission, derived from Equation 2, is shown. In addition, the initial experimental confirmation of the effect is presented in Figure 3. Here, a polarization resolved image of a tungsten ribbon at
2000 K was recorded with a CCD camera looking at 45 degrees through a 700 nm filter. A polarizing beam displacement prism was placed in front of the CCD, allowing for simultaneous recording of p and s components of the ribbon’s image.

Assuming several-percent accuracy, typical for an absolute radiation measurement with semiconductor photo-diodes\textsuperscript{2}, our numerical analysis of Equation 2 and 3 concludes that, for example, the minimal configuration, with two angles, can measure n and k with 20% accuracy, emissivity with 10% and temperature with 5%. The precision of measured values improves with the addition of more angles. Obtaining few-percent accuracy for n and k would require measurements at 10 angles. The unequal spread in error values among n, k, E and T occurs due to their non-linear inter-relation in Equation 2. Note that, in practice, precisely this property has allowed reliable measurements of temperature, while offering only fair accuracy with regard to emissivity\textsuperscript{3}.

The phenomenon of the polarization of emitted and reflected light by the surface of a sample was observed earlier, and has been used in various ways for emissivity correction in various low temperature, 600 K - 2500 K, experiments\textsuperscript{11,12,13}. Notably, most of the published approaches require an external light source, such as a probe laser or a blackbody lamp, and essentially amount to equating (or zeroing) the combined emission from a sample and the reference source by means of the mechanical rotation of optical components. Obviously this approach is not applicable in WDM experiments. Firstly, mechanical rotation times are too long compared to the ns timescales in a typical dynamic WDM experiment; secondly, the temperature in a WDM experiment can be as high as 100,000 K, requiring a very bright reference source, which must be as intense as the thermal emission. To our knowledge, the work of Martin\textsuperscript{14} is the closest to the concept described in this paper. Martin did use the polarization of self-emission at various angles to determine optical constants; however, his setup was based on mechanically rotating components and his measurements were absolute; he did not use the ratio approach (Equation 3). Furthermore, Martin has not extended his technique to dynamic temperature measurements. To our knowledge, the first suggestion of employing polarization phenomena to improve the accuracy of temperature determination in high-temperature WDM experiments appears in a theoretical paper by one of the co-authors in\textsuperscript{15}.

III. Advantages and limitations
Laser polarimetry\textsuperscript{16,17}, also known as ellipsometry, is a commonly used technique to determine the emissivity. Measurement accuracy of a few percent can typically be achieved with this technique. In polarimetry, polarization of the reflected laser beam changes upon reflection from the surface. The measured change in polarization state is connected to the unknown optical constants by the Fresnel formula. In order to discriminate the reflected laser from background thermal light, either a lock-in technique (applied in slow $\mu$s-time-scale experiments) or an intense short-pulse laser is necessary.
As can be seen, polarimetry has the same limitation as the polarization pyrometry: it relies on validity of the Fresnel surface approximation. In order to match polarimetry’s few-percent precision in the optical constants measurement, a polarization pyrometer would require at least 10 angles – a quite elaborate setup, but unavoidable when emissivity measurement is the primary goal. Meanwhile, if the temperature is the primary measurement, then, as mentioned earlier, due to the favorable non-linear relation between temperature and emissivity, a simpler setup with several angles can readily provide an acceptable (for most WDM experiments), few percent accuracy. In this case, the complexity of the technical implementation of the polarization pyrometer becomes less than that of a polarimeter, and comparable to that of a conventional multi-wavelength pyrometer.

Laser polarimetry measures the AC conductivity only at a single wavelength (that of the laser). In polarization pyrometry, measurements at multiple wavelengths can be accommodated rather easily, for example by using a spectrometer as a detector. Such a configuration not only would improve the overall accuracy of the instrument, but would allow for simultaneous measurements of emissivity and AC conductivity at multiple frequencies – an important advantage for WDM research, since the AC conductivity contains valuable information about the electronic band structure of hot material\textsuperscript{15,18}.

In contrast to conventional pyrometry, the polarization-based technique is limited to flat samples and to setups in which enough space is available to accommodate measurements at steep angles. However, by far the most limiting factor is the Fresnel surface condition. If a surface is rough (roughness exceeds $\lambda/10$) or contaminated – a situation typical for samples below melting temperature – then the description of surface optical properties becomes more complex than that in Equation 2. Meanwhile, above the melting-point temperature, the Fresnel formulas are expected to be more valid because contaminants disassociate and surface tension smoothes the surface. Hence the polarization technique is more suitable for WDM experiments. If measurements of a cold and rough surface must be done, the probing $\lambda$ should be shifted towards longer wavelengths – in the ideal case surface roughness must be smaller than $\lambda/10$.

The Fresnel formulas assume a sharp surface boundary. An interpretation of the polarization data, when a surface is expanded requires an emission calculation more complicated\textsuperscript{15} than that in Equation 2. An expanded surface condition occurs, for example, in a partially evaporated sample, in a sample expanding due to hydrodynamic motion or in a release wave of shocked matter. Moreover, due to temperature gradients occurring within the expanded surface region, Kirchhoff’s law (absorption equals emission) is violated, requiring the modification of the Plank formula in Equation 2 – an issue also relevant to conventional pyrometry. Nevertheless, as reported in\textsuperscript{15}, in addition to multi-wavelength measurements,
angle-dependent, polarization-resolved data provides extra information, leading to a more reliable temperature determination.

IV. Proof-of-principle experiment

Design specifications, such as probe angles, wavelength, aligning tolerances, etc., were evaluated numerically using Equation 2. The analysis showed that a setup with three angles, 0°, 45° and 60° (±3° alignment accuracy) that measures emission with 3% accuracy at 900 nm, is sufficient for a reliable demonstration of the concept. Based on these numbers, a two-angle “ratio” prototype (Equation 3) was constructed to enable an initial test of the technique. The objective of the experiment was to apply the polarization pyrometer to a DC-current-heated tungsten ribbon (heated up to 2800 K), and compare the output of the instrument to that of a multi-wavelength pyrometer, which was simultaneously measuring the same area of the ribbon. The ribbon was cleaned and polished to a surface roughness less than few micrometers.

The schematic of the setup is shown in Figure 4. A tungsten ribbon (3 mm x 40 mm x 0.2 mm) and collection optics, probing the same spot (Ø 0.2 mm) at the three angles, are placed in a vacuum chamber (p=1e-6 Torr); the collected light is filtered through 900 nm interference filters and detected by amplified Si pin-photodiodes. An absolutely calibrated spectrometer (Ocean Optics HR4000), looking normally at the ribbon, is used as the reference multi-wavelength pyrometer. Temperature was derived from fitting the continuous spectrum (500 nm-800 nm) to emission models with linear and quadratic dependence of emissivity on wavelength (Figure 5). Corrections for the difference in transmission of p and s channels of the same probe, were done by rotating the sample (mounted on a rotation stage that can be turned without interrupting the current flow through the sample) normally to a probe — a correction coefficient is calculated, assuming that p- and s -radiation must be equal at 0°.

Emissivity and temperature are determined by numerically solving the system of Equation 1 and 3 — first, the emissivity is derived from Equation 3, and then it is used in Equation 1 to solve for the temperature. The measurements were taken at thermal equilibrium for a series of increasing and decreasing filament currents. The temperature, emissivity and optical constants obtained in these series are shown in Figures 6, 7, 8. Error bars are estimated numerically from Equation 3 taking into the account the realistic radiation measurements and alignment errors (the error bars do not include uncertainty due to any failure of validity of the Fresnel surface approximation). Obtained temperatures are compared to those measured with the multi-wavelength pyrometer and to the blackbody temperature, which was calculated from the intensity at normal incidence, assuming an emissivity equal to unity. In addition, the temperature calculated with a tabulated value of emissivity was added to the comparison – an averaged E=0.405 tungsten emissivity from a source² was used to correct the black body temperature recorded with the 0° probe.
VI. Discussion
The constant 0.405 emissivity is a reliable value to estimate the true temperature: according to\(^2\), the 900 nm emissivity decreases from 0.413 to 0.395 – a 4% change relative to the average – when the temperature is increased from 1200 K to 2200 K. Due to the non-linear relation in Equation 1, the 4% emissivity variation would manifest in less than a percent temperature variation, which is less than the expected overall accuracy of the instrument. Note that the emissivity in other published references agrees with that in\(^2\) within 5%. For clean comparison, it is worth mentioning that tabulated values of emissivity are usually obtained with techniques other than the polarization method. The emissivity is commonly measured via either laser reflectivity combined with integration spheres, or the blackbody-cavity comparison technique\(^{19,20}\).

In Figure 6, between the multi-wavelength- and polarization-derived temperatures, the latter has a far better agreement with the table-derived values. This serves as the first evidence that in the described experiment, the polarization technique worked and performed better than a conventional multi-wavelength pyrometer. The noticeable discrepancy of the multi-wavelength temperature originates from the false square law assumption: the emissivity of tungsten in the spectral range, although smooth, has a behavior more complex than a square law\(^2\). Remarkably, despite this, the curve fits in Figure are flawless. Here, another drawback of the multi-wavelength method is obvious: the quality of the curve fit cannot be a reliable measure of the temperature accuracy; without having any knowledge about the emissivity and simply judging by the nearly perfect fit alone, one might have falsely assumed a high accuracy of the derived temperature.

In Figure 7, the emissivity measured with both pyrometers decreases with temperature monotonically— a trend that has been also observed in the literature\(^2\).\(^{19}\) Absolute values of emissivity measured with the polarization pyrometer match the reference\(^2\) better than those measured with the multi-wavelength pyrometer. Still, noticeable disagreement between tabulated data and the polarization-derived emissivity can be explained by the incomplete validity of the Fresnel approximation assumed during the data processing.

Finally, trustworthy information on the optical constants of tungsten is less available than that on emissivity. Publication-to-publication variations in constants' values were found to be as large as 300% at room temperature (Figure 8), with even larger inconsistencies at higher temperatures – these differences may be a consequence of different sample preparation or surface properties. As there is no "reliable" reference, the derived n and k are compared to room temperature values found in\(^{21,22}\).

VII. Conclusion
The polarization pyrometer has the potential to significantly improve temperature measurements in WDM experiments. It can supplement multi-wavelength
measurements, or serve as a stand-alone system. Based on our test results, we conclude that the polarization pyrometer offers far better agreement, both in temperature and emissivity, with tabulated references than a multi-wavelength pyrometer. Successful agreement of temperature measurements in the static experiments encourages and justifies the next step in the validation of this diagnostic, for dynamic WDM experiments at higher temperatures.

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Figure 1: Concept of the polarization pyrometer: thermal emission from a sample is collected at several angles, split into two polarization components with polarization optics (polarizing cube, Wollaston prism, thin-film beam splitters, etc), filtered spectrally, and focused on a photo detector.
Figure 2 Simulated angular dependence of p- and s-polarized thermal emission of tungsten-like (n=3.6, k=2.9) sample at T=2500 K, λ=900 nm.
Figure 3: Emission of a DC-current-heated tungsten strip as seen by a CCD camera through a polarization beam splitter (a polarizing beam displacement prism) at 45 deg, 700 nm. Image has been corrected to reflect the true radiation ratio.
Figure 4: Setup for the static proof-of-principle experiments. Each probe consists of a relay lens system, L₁ (f=150 mm, Ø25 mm) and L₂ (f=30 mm, Ø25 mm). The resulting 5x magnification optics relays the Ø 0.2 mm fiber core to a Ø 1mm probing area (shape of 45° and 60° probe is an ellipse). Polarization resolution was performed at the earliest possible stage, in the vacuum right after the first uncoated lens, L₁. In the 45° and 60° probes, collimated light was split into two beams (legs) with polarizing beam splitting cubes (1000:1 extinction ratio). The collected light was coupled to a multimode-optical Si-fiber (5 m long, Ø200 µm core, na=0.2), which transmitted light via a feed-through to a photo-diode outside the chamber. The thermal radiation was filtered by a 900 nm (40 nm transmission) interference filter and detected by an amplified (DC-300MHz) Si pin-photodiode. The 0° probe was calibrated absolutely with a tungsten ribbon lamp, a NIST traceable calibration standard.
Figure 5: Thermal, spectral density recorded by the Ocean Optics spectrometer and best fits to emission models with linear and quadratic dependence of emissivity on wavelengths. Temperatures obtained from the fit are 2170 K and 2110 K correspondingly. Fitted curves are close to each other and almost indistinguishable in the plot. Spectral data was taken for each data point in Figures 6, 7, and 8.
Figure 6: Comparison of temperature measured with polarization pyrometer and multi-channel pyrometer. Solid circle – polarization pyrometer; stars – table emissivity; square – quadratic emissivity; gray circle – black body temperature ($\varepsilon=1$).
Figure 7: Comparison of emissivity obtained with polarization pyrometer and multi-channel pyrometer. Solid circles – polarization pyrometer, squares – quadratic emissivity, solid curve – emissivity of tungsten at 900 nm from²
Figure 8: Optical constants at 900 nm determined from the polarization pyrometer measurement. Squares – refraction coefficient, n; diamonds – extinction coefficient, k. Solid lines are optical constants of tungsten at room temperature, at 900 nm from\textsuperscript{21,22}.

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