

THERMAL INFRARED REMOTE SENSING AND KIRCHHOFF'S LAW: I
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With the near-term prospect of thermal infrared spectroscopic measurements of martian surface materials by Mars Observer, it has become important to understand the emissivity behavior of materials, especially particulate materials. Perhaps the most fundamental question to be answered is the extent to which such materials follow Kirchhoff's Law [1] under less than ideal conditions.

Kirchhoff's Law is typically stated in its simplest form (for opaque materials without wavelength or directional subscripts) as $E=1-R$, where E and R are emissivity and reflectance, respectively. However, this Law, as originally conceived, applies only to samples in thermal equilibrium with their surroundings-- i.e., the sample is isothermal and at the same temperature as the background to which it radiates. Most laboratory measurements of emissivity only approach this condition and it never applies in remote sensing applications. In particular, the background is often much cooler than the radiating sample, and this has led to a long controversy about the applicability of Kirchhoff's Law under such conditions. It has also led to field and laboratory emissivity measurement techniques that use some form of the "emissivity box" approach, which surrounds the sample with a background as close as possible to the sample temperature [2,3]. However, clear theoretical arguments have been made that a "freely radiating" sample (i.e. sample independent of an environmental radiation field) will still follow Kirchhoff's Law, if the sample material obeys the Boltzmann distribution [4], resulting in Planckian emission.

However, non-Planckian emission behavior can occur in particulate materials when a thermal gradient is induced and the bulk sample no longer obeys the Boltzmann distribution. This is best illustrated for remote sensing purposes by laboratory measurements of the spectral emissivities of fine particulate samples under simulated lunar environmental conditions [5]. Here, the lack of interstitial gas molecules results in very slow heat transfer between grains by conduction across the small asperity points of contact, compared to the more rapid convective heat transport when gas is present. Under such conditions, a cold skin of particles can develop over a warmer interior (the solid state greenhouse effect) [5]. The net result is increased spectral contrast and distortion of the spectral curve compared to directional hemispherical reflectance measurements. In short, Kirchhoff's Law does not apply because of the non-Planckian source function of the emitted radiation. Although the thermal gradient is less severe in a fine particulate sample under terrestrial or even martian conditions, where interstitial gas provides convective heat transport, no measurements have been made to determine the exact extent to which Kirchhoff's Law may still be applied. Thus, there is uncertainty about the extent to which laboratory measurements of directional hemispherical reflectance can be used to predict emissivity simply because of the presence of thermal gradients.

Theoreticians are fond of using simple examples in their arguments, such as large solid surfaces, where geometrical optics hold and scattering is not important [4]. However, geometrical optics do not hold for particulate samples with grain size $\leq \lambda$, and absorption efficiencies in this case can exceed unity [6], implying a possible violation of Kirchhoff's Law. Further, individual particles in a geological sample may

have a highly complex interaction with radiation. The ultimate in complex samples may be soils in which the larger, opaque (in the reststrahlen bands) quartz grains are coated with a semi-transparent layer of tiny, optically thin particles of clay and quartz. The particulate nature of these soils enhances scattering, and the combination of opaque grains with semi-transparent coatings results in an extremely complex radiative transfer problem. Such particulate samples are also more likely than solid samples to develop thermal gradients when radiating to a cooler background, due to their low thermal conductivity. Thus, such samples provide a severe test of Kirchhoff's Law.

In our initial experiments, we have heated soil samples in air on a hot plate in the laboratory to a much higher temperature ($\sim 50^\circ\text{C}$) than the room temperature background. Spectral emissivity was measured as described in [7], except the known emissivities (determined from reflectance) of both the primary and secondary Christiansen features were used, instead of assuming an emissivity of unity at these wavelengths. This measured emissivity was compared to directional hemispherical reflectance measured as described in [8]. As shown in the figure below, spectral emissivity calculated from reflectance is very close to measured emissivity, despite noise in the emissivity curve due primarily to atmospheric water vapor. In fact, measured and calculated emissivity agree within better than 1%, which is within our estimated experimental error of 2% with such noisy emission data obtained under high humidity conditions. Thus, despite the complexity of the sample and the marked temperature difference between sample and background, Kirchhoff's Law can still be used to predict emissivity from laboratory measurements of reflectance.

A subsequent paper will address similar comparisons made in the field under realistic remote sensing conditions.

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