Terahertz

Executive Summary

Here, we extend our use of ultrashort laser pulses as a means of generating intense THz-wavelength pulses as a means of potential stimulation of AHE. These intense pulses are further enhanced by the implementation of PdD_x phonon-resonant split-ring structures that act as microscale antennas. Ultimately, we find that the electron-driven optical properties of metallic PdD_x causes screening that well exceeds the potential interaction of light with the phonon mode of interest.

Stimulating optical phonons for AHE

A non-negligible fraction of LENR/AHE literature discusses the importance of phonons, or vibrations of atoms, within palladium deuteride toward generating anomalous heat events. It is worth noting that such literature is relatively qualitative in nature and does not provide mathematical rigor to such "theories," but, nonetheless, we seek to leave no stone unturned. The difficulty with the lack of rigor in these discussions is that there is negligible work in the LENR community that is definitively stimulating these atomic vibrations of interest within PdD_x. In other words, many authors cite the importance of these phonons, but their methods do not stimulate the phonon modes!

This limitation is likely due to the fact that there are very few ways of "coupling" to phonons in a material system, particularly metals. As described in our USP section, we tested this theory by "beating" the sample via repetitive short pulse heating at the resonant frequency associated with the Pd-D optical phonon mode centered at ~14 THz. While this qualitatively may be expected to increase the amplitude of the phonon, it certainly is not a "known" method in literature for phonon stimulation.

In fact, the only known method of directly interacting with and manipulating phonons at this frequency of interest is through optical stimulation, where the frequency of the *photons* are resonant with the frequency of the *phonon*/lattice vibration. This is fundamentally different than our above resonant heating approach: In the resonant heating approach in USP, we simply "hit" the material repeatedly at 14 THz. However, this mechanical driving force is simply not equivalent to a direct interaction with atoms themselves. In contrast, when light has a frequency or wavelength resonant with the atomic vibration, direct coupling can be directly observed; this is the basis for a number of spectroscopy methods such as FTIR (Fourier Transform Infrared

Spectroscopy) and has been shown in multiple studies to provide a manipulation of phonons, including an increase in their amplitude[34].

In the following sections, we discuss our attempts at using resonant-frequency optical pulses (THz pulses) to stimulate AHE events. This includes the development of intense THz sources, measurements on the coupling between these sources and loaded Pd, as well as the development of resonating structures to enhance the electric field intensity experienced by the PdD_x.

Developing intense THz sources

The primary method of generating *intense* THz sources is via non-linear processes that occur when an intense, ultrashort pulse from a laser interacts with a gas or liquid. Consider the electric field amplitude of a laser pulse is given by $E = \sqrt{P * 2\eta}$, where P is the power flux of the laser source and η is the resistivity of free space. Thus, to generate a sufficient electric field, E, for nonlinear processes (nonlinear processes typically have susceptibilities 6-8 orders of magnitude lower than linear susceptibilities), the required power increases with increasing field intensity (i.e., more power is needed to drive small changes in E when E is large than when E is small – there are diminishing returns!). This limitation suggests that extremely large powers are required for large electric fields. For this reason, ultrashort pulse lasers become an ideal "driving" function for the electric fields of interest; ultrashort pulse lasers deliver their total energy in extremely short periods of time. Since the power follows $P = \frac{Energy}{\tau_p}$, where τ_p is the duration of the pulse, the *shorter* the pulse duration, the greater the power, even for the same net energy input. There are no other methods of generating equivalent power densities; for example, a factor of $\sim 10^{12}$ is gained by relying on a picosecond pulse rather than a 1 second pulse duration. The relative intensity of the pulses used in this work produces *peak* powers of nearly 100 GW, the equivalent of nearly fifty large-scale nuclear reactors.

Qualitatively, this process of THz generation via ultrashort pulse lasers functions by the extremely high electric field of the pulse driving electrons out of the molecules in a gas/liquid; an example image of this plasma-forming process is shown below in Fig. 1. Notably, in the absence of the graphical depiction of the laser and corresponding THz emission, both beams are undetectable by eye or with standard cameras. The ionized gas/plasma is visible due to the large number of wavelengths emitted from this ionization process; these wavelengths span into the THz region under appropriate conditions.

[34] Woerner, M., *et al., Physical Review Letters* **122**, 107402 (2019).

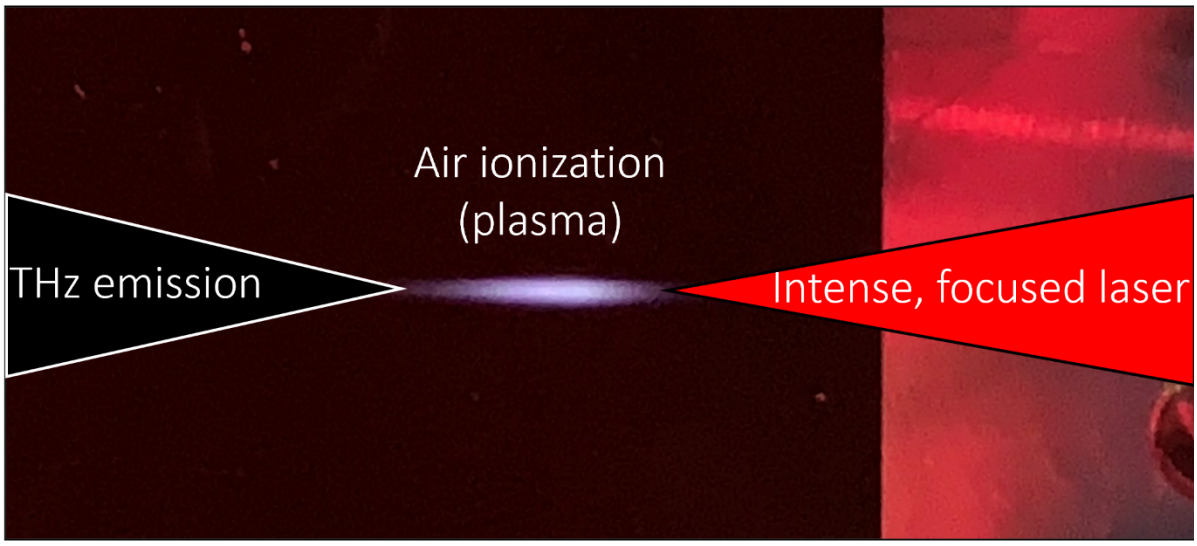

Figure 1: Experimental image of laser-induced ionization for THz pulse emission.

Due to the large wavelength of THz pulses, there are few methods available for *detecting* the generation of these pulses. As mentioned above, standard cameras do not detect these wavelengths of light; in fact, even specialty electronics specifically designed for "far wavelength" operation do not generally reach the frequencies of interest. The primary method for detecting infrared light of these wavelengths is via interferometry, where the infrared light is split into two paths and the constructive/destructive interference is measured as a function of path length (this is the principle of FTIR discussed later). While this works sufficiently for *continuous* waves of light, the method struggles at the low duty cycles for intense, quasi-single cycle THz pulses of interest here. Rather, we rely on a "pump-probe" method of detecting the generated THz pulse.

Our method of detecting these THz pulses is a four-wave mixing process referred to as Air-Biased Coherent Detection (ABCD)[35]. As we use an 800 nm pulse to *generate* the THz wave, we can also use the pulse to *detect* the THz wave. This is done by splitting off a small, ~10% fraction of the 800 nm pulse prior to generating the air-plasma. Once the air-plasma is generated, we use Si optics to remove the remnant 800 nm beam within the THz beam path; this isolates our THz "pump" beam to wavelengths more than ~1200 nm, thus eliminating the visible components of the air-plasma as observed in Fig. 1. From this, we recombine the 10% "probe" beam at 800 nm and the THz "pump" beam such that they interact with each other spatially and temporally; by varying the temporal overlap of the two beams, we can extract the temporal profile of the THz pulse by reading-out the second harmonic of the 800 nm probe as a function of time-delay.

Quantitatively, this ABCD process is dictated by the following equation,

$$E_{2\omega}(t) \propto \chi^{(3)} E_{THz}(t) E_{\omega}(t) E_{\omega}(t),$$

[35] *X. Lu and X.-C. Zhang, Applied Physics Letters* **98**, 151111 (2011).

where $\chi^{(3)}$ is the third-order susceptibility of the detection medium (here, the medium is air), E_ω is the electric field of the fundamental probe wavelength (here, ω = c/800 nm), E_{THz} is the generated THz pulse, and $E_{2\omega}(t)$ is the generated second harmonic via four-wave mixing of the two electric fields (here, 2ω = c/400 nm). Notably, this detection is simply the inverse of the generation process which follows,

$$E_{THz}(t) \propto \chi^{(3)} E_{2\omega}(t) E_\omega(t) E_\omega(t),$$

as we also rely on the second harmonic generation of our 800 nm generation beam to create a sufficiently intense THz pulse.

An example of this time-domain detection method is shown below in Fig. 2. As shown, the second harmonic signal displays various oscillatory features within the electric field associated with the THz pulse. While this is our primary information of interest, it is inconvenient for understanding the resultant pulse. Rather, we can perform a Fourier-transform on this time-domain signal to extract the frequencies contained within the pulse; this is shown on the bottom of Fig. 2. This displays the wide range of frequencies contained within the THz pulse; our goal is to generate *intense* peaks as close as possible to 10-14 THz. In the example shown here, we can see that the highest nearby frequency peak is around 8 THz and corresponds to the most intense peak within the upper waveform.

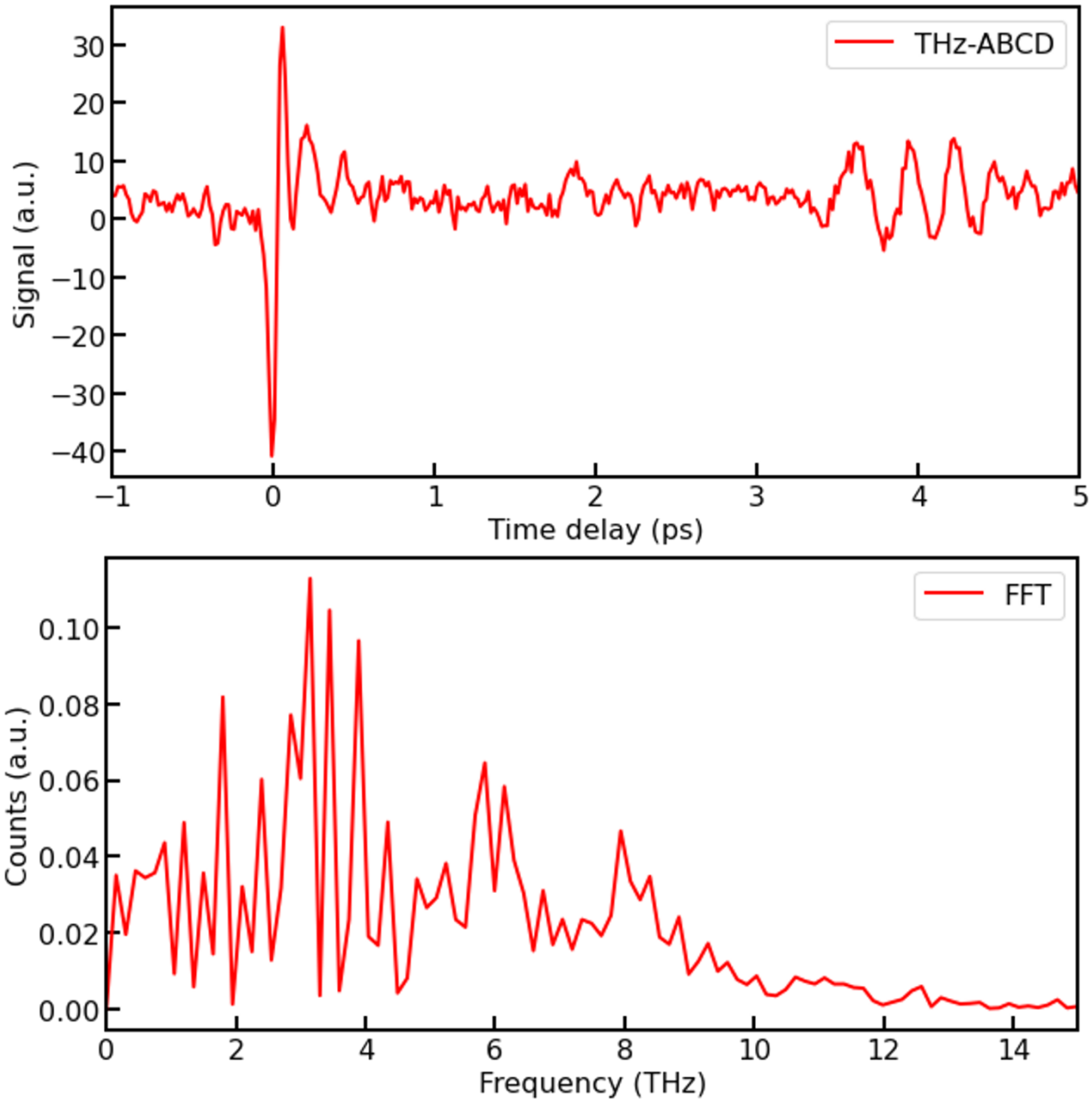

Figure 2: (Top) Representative ABCD signal produced via four-wave mixing of the generated THz pulse with an additional 800 nm "probe" beam. (Bottom) Fourier-Transform of the upper ABCD waveform, providing a clearer representation of the produced frequencies.

We estimate the peak electric field based on several experimentally measured properties of the pulse. The peak electric field for a THz pulse is given by,

$$\left|E_{peak}\right|^2 \approx \frac{2W\eta_0}{\tau A},$$

where W is the pulse energy, η_0 is the impedance of free space, τ is the pulse width, and A is the beam area. The pulse width can be extracted through the ABCD waveform, e.g., as shown in Fig. 2. The beam size is determined using a THz Beam Profiler (WinCamD-THz); we measure the pulse width at the beam waist. The pulse energy is given by bolometry measurements of the produced beam. These measurements, when performed in a dry atmosphere (water molecules have strong absorption bands in this frequency range, greatly reducing the beam intensity), suggest field intensities on the order of 500 – 1000 kV cm^{-1}.

Resonators

One approach to further increase the electric field experienced by a material is via beam interactions with a metamaterial. For example, radio waves rely on antennas for enhancement of the electric field and optical waves can interact with plasmon waves within a metal for enhancement of the electric field. In the former case, antennas are generally developed toa maximum frequency of about 0.1 THz, beyond which resonant structures become significantly more difficult to create. On the other end of the spectrum, metal plasmons and other polaritonic modes are generally supported from 100s of THz down to about 30 THz, where, again, a gap is observed. This range, 0.1 – 30 THz, is referred to as the "THz gap," as useful power generation and receivers (antennas) are unfeasible.

While a large amount of research has been performed to reduce this gap, there remains difficulties in resonant enhancement of the electric field. To address this and further increase the electric field intensity of our THz wave, we investigated the production of split-ring resonators[36] that specifically enhanced the frequency range of interest (10-14 THz).

To optimize the design of these structures, finite element calculations were performed on a large variety of metastructures; these calculations were guided by electromagnetic constant measurements of Pd/PdD_x and other materials of interest (e.g., the dielectric substrate, gap materials, etc.). Further, we performed measurements post-fabrication to verify these calculations. An example of these finite element calculations is shown in Fig. 3 below, where the color-axis displays the normalized electric field (electric field / incident electric field) at the resonant frequency. Additionally, an example of the idealized design for 10-14 THz enhancement is shown in Fig. 3, where we determined that *double* split-ring resonators (dSRRs) led to even greater enhancements in the electric field. Ultimately, our simulations suggest that the electric field within the gap of these dSRRs is of order 100-1000x the incident electric field intensity; these resonators are made from Pd to utilize it as a potential stimulation material.

[36] Driscoll *et al., Applied Physics Letters* **91**, 062511 (2007).

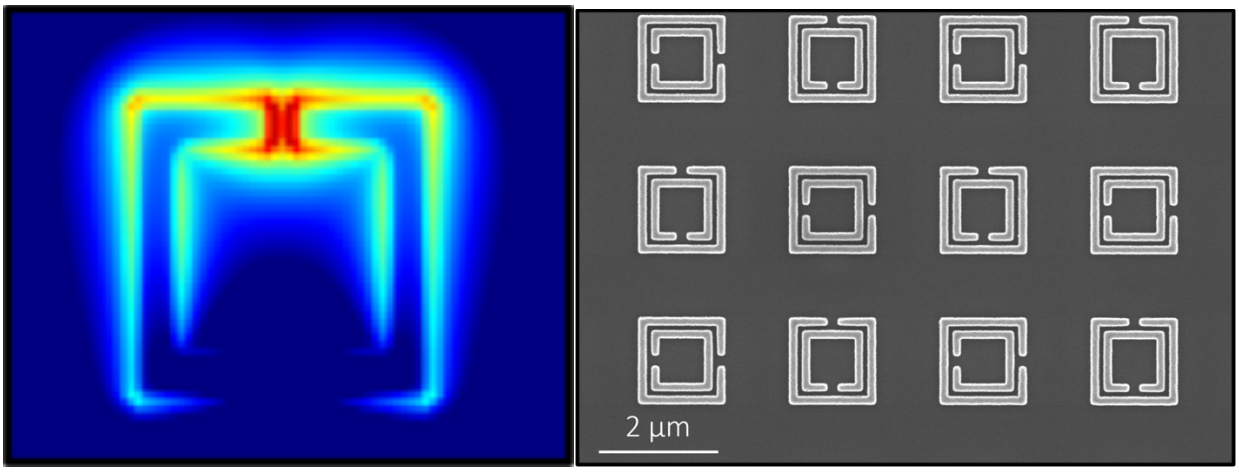

Figure 3: (Left) Finite element calculations of a single split ring resonator. The color axis is the normalized electric field, where red denotes enhancements in the electric field and blue denotes no enhancement. Most of the field enhancement, as expected, occurs within the "split" of these structures. (Right) Double split-ring resonators designed and built by us for improved enhancement in the 10-14 THz wavelength range.

To further highlight the benefit of these structures, we show below, in Fig. 4, a direct comparison between various resonator structures, including no resonator, a single split ring, and split rings with offset gaps (i.e., not at the midpoint of a side). We experimentally measure the *transmission* of light through these structures to determine the resonant enhancement: The absorption of light is directly proportional to the "coupling" between the electromagnetic wave and the structure of interest. Coupling in excess of a "non-resonant" structure is direct verification of electric field enhancements. Further, our simulations provide not only enhancement, but an expected transmission function: the simulated transmission can be directly compared to our experimental measurements to allow for improved quantification of this enhancement.

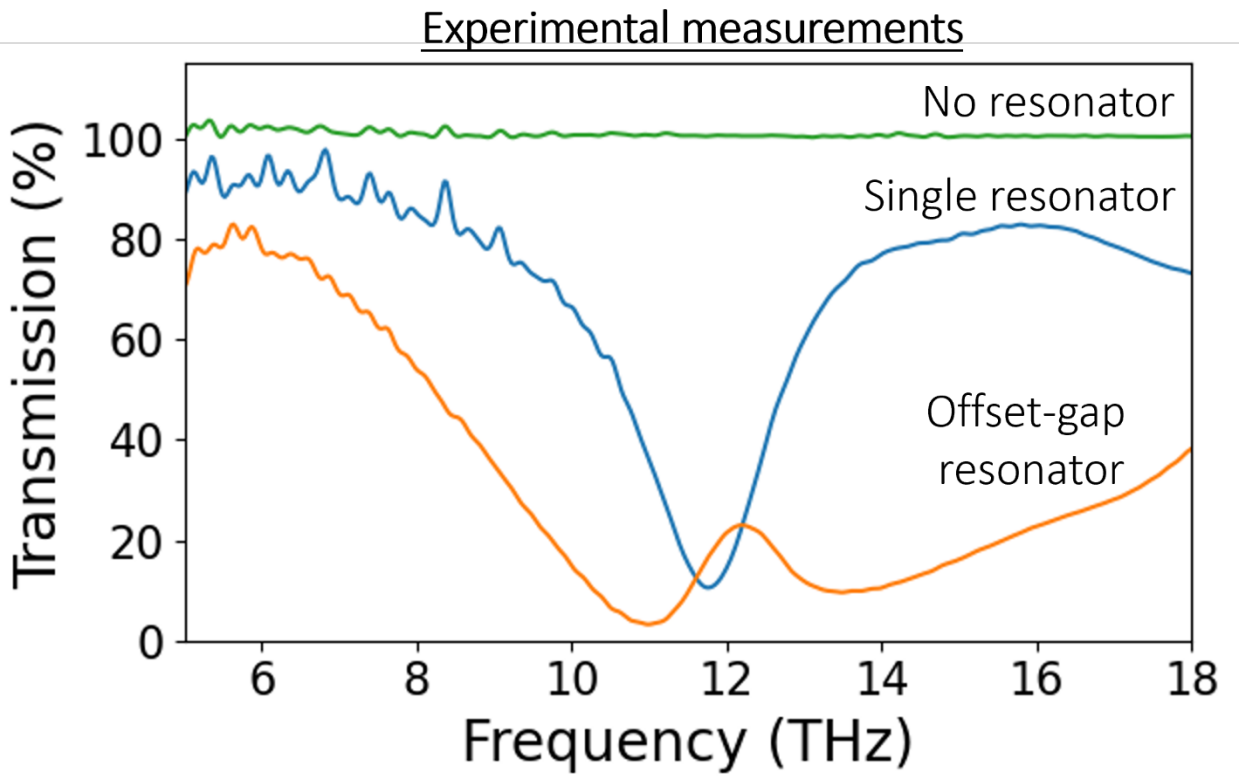

Figure 4: Experimental FTIR measurements on (green) Si with no resonator, (blue) Si substrate with a single split ring made of Pd, and (orange) an offset-gap designed Pd resonator on a Si substrate.

Notably, we performed THz pulse stimulation on structures with various resonant frequencies/materials to look for AHE events via the production of "hole" events within the metamaterial. These experiments were performed by sweeping/rastering the THz beam over a wafer containing a large number of these dSRR structures, as well as moving the beam through the focal plane (i.e., performing this experiment at repeated distances from the expected beam focus) to ensure that the beam reached maximum intensity. Ultimately, **we did not observe laser-induced nor AHE-induced hole/void formation within these dSRRs using the intense THz pulses described above**. We believe that there is simply insufficient coupling between the light and metal of interest (PdD_x); this notion is well-supported by literature and internal measurements on PdD_x and discussed in more depth below.

Electromagnetic coupling to phonons in metals

While THz and mid-infrared coupling of light into *dielectric* materials is well-known, predictable, and commonly studied, there is a large lack of literature on the coupling of mid-infrared light to

phonons within *metals* such as Pd. Generally, this is not surprising as the dielectric function that describes light-matter interactions in metals follows the Drude model. While the Drude model can support resonant interactions with, in theory, an oscillatory mode, one would not expect this to occur at energies *below* the plasma frequency; in fact, this limitation is the reason why metals do not support strong coupling in the mid-infrared and researchers have turned to dielectrics!

The Drude model is derived from kinetic theory (quasi-classical calculations) that assumes atoms within a material are "rigid" and simply act as stationary scattering sites for the electrons that cause electrical conduction (and reflectivity) within metals. Notably, this simple model provides *extremely* good agreement with the optical properties of metals at wavelengths far from *electronic* transitions. The optical properties of a metal follows the Drude dielectric function, ϵ, given by,

$$\epsilon(\omega) = \epsilon_0 \left(1 - \frac{\omega_p^2}{\omega^2 + i2\omega\gamma}\right),$$

where ϵ_0 is a constant representing the dielectric function at infinite frequencies, ω_p is the plasma frequency (the frequency in which electrons stop responding to light: this is generally at deep UV wavelengths for metals), and γ is the scattering rate of electrons due to the rigid lattice. This function is notoriously smooth and leads to the extremely high reflectances of metals in infrared and greater wavelengths (i.e., THz wavelengths).

The key point, is simply that the optical properties of metals such as Pd/PdD_x are well-described by a model that invokes *no* consideration of the phonons other than their role on free-electron scattering; the interaction of light with metals at THz wavelengths is dictated by electrons, *not* phonons of interest for our AHE studies. We confirmed this by a number of FTIR experiments on monitoring the absorption of THz/mid-infrared light with PdD_x and PdH_x – in no cases did we observe absorption due to phonon modes of interest. This makes electromagnetic stimulation a non-ideal method for testing the phonon hypothesis of driving AHE; we explore alternative approaches to this hypothesis in our USP effort.