Reply to "Comment on 'Evidence of a first-order phase transition to metallic hydrogen' "

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Goncharov and Geballe reanalyze [Phys. Rev. B **96**, 157101 (2017)] experimental data using a finite element analysis and cannot reproduce our experimental heating curves; they use incomplete and incorrect material properties. They analyze optical data at the onset of the insulator-to-metal transition to show that it cannot be fit to a Drude model, not realizing that the Drude model does not describe metallic hydrogen in this transition region. Yet, they ignore the region where the reflectance is saturated and the data can be fit to the Drude model. They suggest that we observe semiconducting behavior rather than metallic behavior, but a straightforward calculation shows that reflectance from a semiconductor cannot explain our observed reflectance.

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In our experiment, molecular hydrogen at static high pressures (P's) is sandwiched between two diamond anvils; one of the anvils has a thin tungsten film that is semitransparent. This film absorbs ∼30–40% of the heating pulsed laser light and warms to high temperatures (T's), heating the hydrogen in contact with its surface. The semitransparent film enables us to measure both transmission and reflectance of the hydrogen as a function of photon energy using time-resolved spectroscopy. We measure heating curves (Fig. SI5 of our paper), that is, the temperature of the tungsten film as a function of laser power. At certain values of P,T we observe plateaus that are interpreted as arising from the heat of transformation for a phase transition to metallic hydrogen (MH). These values of P_c/T_c for the pressure dependence of the transition are in reasonable agreement with the latest quantum Monte Carlo calculations [1] for this liquid-liquid phase transition, also known as the plasma phase transition (PPT). With increasing laser power the temperature rises above the plateau temperature, and the measured reflectance rises until it saturates at a value of 52–55% (Fig. 4 of our paper [2]), measured for optical wavelengths that vary by a factor of ∼2 (514–980 nm). When the reflectance is saturated, the transmission is negligible (Fig. 4 [2]), and this is considered the bulk MH region. This is the expected behavior of a metallic film as it is created and thickens: The transmission goes to zero, and the reflectance goes to the bulk value.

In analyzing our optical data, Goncharov and Geballe (GG) [3] use a Drude free-electron model but evidently are not aware that the Drude model is not applicable at the onset of the transition where the reflectance and absorption are low, according to theoretical models [4,5]. In Fig. 3 of their Comment, they cannot fit to a Drude model in this region as a critique that our sample is not metallic. They actually find what the theory predicts that the Drude free-electron model is not applicable in this region. Had they fitted in the saturated region, they would have found an excellent fit of the energy-dependent reflectance to the Drude model as we have found [Fig. 1(a), this Reply]. From this fit, one can determine a complex dielectric function of a metal [6]. Using the Drude model in the saturated region at the highest pressures, we find the static electrical conductivity is 15 500 ± 1500 S/cm, whereas the plasma frequency is 21.8 ± 2.8 eV. This value of the conductivity is much higher than the Mott-Ioffe-Regal (MIR) criterion for a liquid metal and is in agreement with theoretical values found at similar conditions [4,7]. Thus, this objection of GG to our optical data is not supported by their analysis.

GG state that the transition points P_c/T_c of the PPT and the critical points are contradictory. The existence of a critical point for the PPT is common to all theories of this phase transition. At present there are no experimental determinations of the critical point; thus, no contradiction. A further criticism of their optical analysis, shown in their Fig. 3, is that they assume a scattering time τ of $1.4. \times 10^{-17}$ s. The MIR minimum metallic conductivity criterion is based on the minimum scattering length being the average separation between atoms. Their value of τ yields a scattering length of 0.26, the average atomic spacing, which is unphysical.

GG suggest that our sample is semiconducting with a continuous closing of the energy band gap. In our experiment the transition to the metallic state is abrupt with no observable concomitant interband transitions in the frequency range investigated. In Fig. 1(b) we show the expected reflectance for a thick degenerate semiconducting sample of hydrogen with a 0.2-eV band gap (the details are discussed elsewhere [6]). The large disagreement between the reflectance expected for semiconducting hydrogen (∼0.17) and our observed data (∼0.55, Fig. 4 [2]) allows us to rule out thermal smearing of a reduced mobility gap as has been suggested by GG and one of the authors of the Comment, elsewhere [8].

GG also perform a finite element analysis (FEA) of heating curves. An FEA should be designed to explain the experimental data; they present a model to obtain a result that does not simulate our experiment. In their FEA they do not include the possibility of the formation of MH as seen in their table where they only give material values for molecular hydrogen (*Note*. The specific heat should at least double in value for full dissociation as the number of particles double). They fail to reproduce the observed plateaus in our heating curves (Fig. 2 in their Comment) and rather show a change in slope, which is in stark contrast to our data. To accomplish this fit they use absorption data from an experiment by McWilliams *et al.* [8] which has been shown to be flawed due to the experimental geometry [9]. The thermal conductivity they

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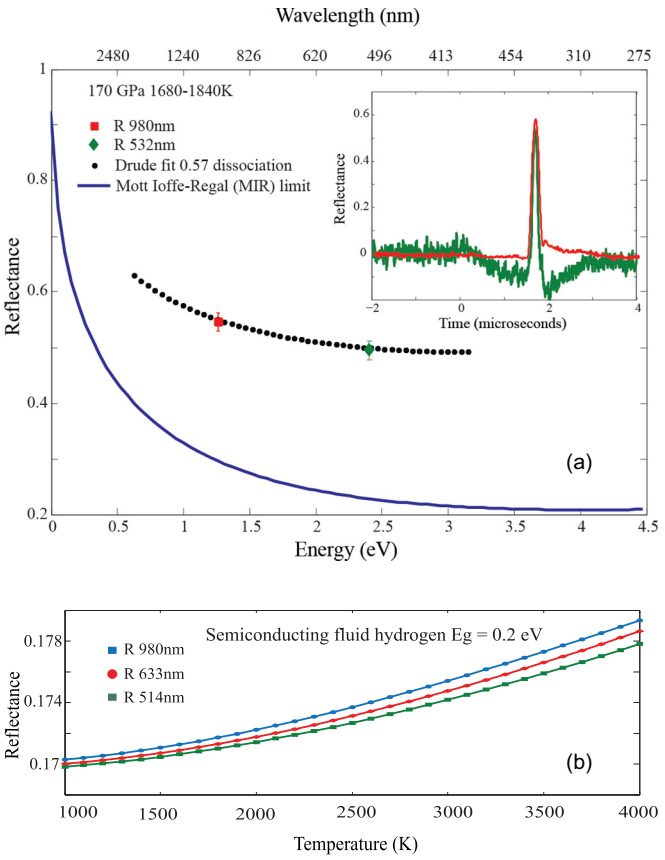

FIG. 1. (a) Bulk liquid metallic hydrogen (LMH) reflectance as a function of energy or wavelength, plotted for 170 GPa. The Drude-free electron fit to the experimental data is derived from a least-squares fit to the energy dependence of the measured reflectance. The inset shows time-resolved reflectance measurements. (b) Expected reflectance of dense hydrogen due to thermally activated carriers, plotted against temperature if hydrogen was semiconducting with a 0.2-eV band gap. This result is for a thick nontransmitting sample to be compared to a similar sample of metallic hydrogen (Fig. 4 [2]).

used for molecular hydrogen in their simulations is rather high, 10–100 W/mK, values typical of conducting metals. Moreover, this value is also a factor of 2–20 higher than the value previously used by one of the author's own FEA analyses of hydrogen (see Table IV of Ref. [10]).

GG analyze the thickness of our sample and state "*increases in reflection of up to 13% in the plateau region are interpreted as being due to the transformation of thin layers*". They use their FEA model with guessed material properties to determine a sample thickness of ∼200 nm. By contrast, the experimental dielectric function derived from the energy dependence of our optical reflectance measurements allows an experimental determination of the thickness of the metallic hydrogen film, shown in Fig. 2. This clearly demonstrates that, as the transmission goes to zero, the thickness of the metallic hydrogen layer is on the order of tens not hundreds of nanometers. From our determination of the plasma frequency using data from the region of saturated reflectance, we find a degree of dissociation of ∼57%, whereas in their analysis full dissociation is assumed, which is clearly not warranted by the data.

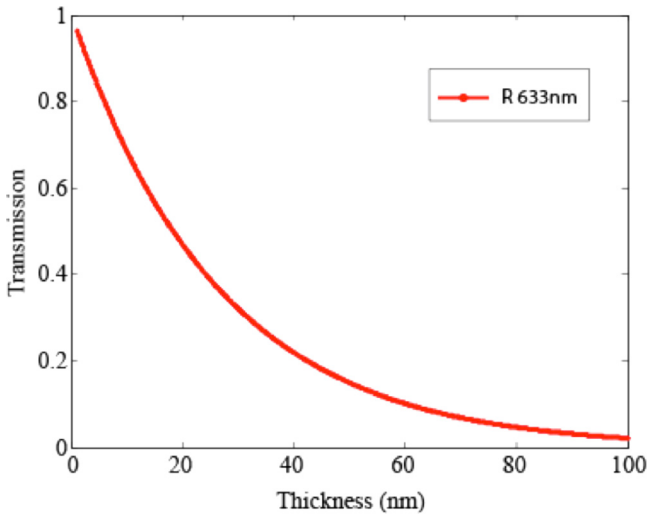

FIG. 2. Transmittance of metallic hydrogen layers as a function of thickness plotted for 633-nm light.

Finally, GG use a Drude model in a region where it is not applicable [9] to determine a static conductivity of 590 S/cm. From our measurements in the region where the Drude free-electron model is applicable, we find values of ∼10 000–15 000 S/cm.

To summarize, there are no grounds for the putative inconsistencies of our observation of LMH as claimed by GG. We reiterate that the observed saturation of high reflectance (∼50%) and the Drude-like energy dependence of this reflectance clearly reveal a highly conducting state with a high carrier density. The high-temperature phase of hydrogen is thus conducting, degenerate, and largely dissociated. All of these are qualities of a metallic fluid.

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