

Experimental and calculational equation of state and melting curve of CeH₂₋₃

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Diamond anvil cell synchrotron x-ray diffraction experiments, molecular dynamics, density functional theory, and Lindemann melting theory were used to investigate the compressibility and melting properties of CeH₂₋₃. Ambient-temperature synchrotron x-ray diffraction measurements were made in diamond anvil cells up to a peak pressure of 20 GPa and show no indication of a first-order phase transition over this pressure range. Equation-of-state fits to the diamond anvil cell data constrain the isothermal bulk modulus, B_0 , to the range 76.7(51)–87.2(39) GPa. Molecular dynamics and density functional theory were employed to simulate compression curves to a peak pressure of ~ 30 GPa, and constrain B_0 to the range 71.6(12)–81.9(12) GPa. Using laser-heated diamond anvil cells, five melt points were measured at pressures between roughly 2 and 10 GPa. We report an experimentally determined zero-pressure melting temperature of 1737(75) K and $dT/dP = 27(15)$ K/GPa. A high-pressure melting curve generated from molecular dynamics simulations shows linear behavior between 0.5 GPa and the peak simulated pressure of 5 GPa, with a fit slope of $dT/dP = 16$ K/GPa over this range. Calculations based on Lindemann melting theory yield a zero-pressure melting temperature of 1609 K and a slope of $dT/dP = 44$ K/GPa for CeH₂.

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I. INTRODUCTION

Cerium hydrides and deuterides have been studied for nearly a century, with the first x-ray diffraction (XRD) measurements reported in 1934 [1]. Recent interest in cerium hydrides has focused on reactive transport modeling of shock ejecta [2,3] and investigations into the high-pressure and high-temperature structures of potentially superconducting “superhydride” or “polyhydride” compositions [4–7]. Early neutron scattering and spectroscopic experiments determined phonon dispersion curves, explored compositional effects on crystal field excitations, analyzed the formation and composition of hydride growth centers, and calculated force constants of CeH₂₋₃ [8–11]. A complementary suite of computational studies have employed density functional theory (DFT) and molecular dynamics (MD) to simulate fundamental physical properties of cerium hydrides (e.g., phase stability, elasticity, magnetic properties, liquid structure) under various pressure and temperature conditions [12–15].

The compositional phase diagram and structural properties of cerium hydride under ambient pressure and low to high temperatures are now well understood through XRD and calorimetry experiments [1,16–21]. At room temperature and

ambient to moderate pressures, cerium hydrides crystallize mostly in the cubic system as CeH₂₋₃, with the exception that CeH_{2.28-2.64} crystallizes in a tetragonal structure [20]. The unit cell volume of cerium hydride initially expands as hydrogen is introduced into the face-centered-cubic (fcc, γ phase) Ce metal crystal lattice up to the composition CeH₂. However, the lattice volume of cerium hydride then contracts with increasing hydrogen content for compositions ranging from CeH₂ to CeH₃ [20].

Despite more than 90 years of research, including high-pressure and high-temperature experiments and simulations, the equation of state (EOS) of cerium hydride remains poorly constrained. Existing experimental pressure-volume data access high pressures but are sparse and show considerable scatter in volume [4–7]. Furthermore, previous reports on the melting temperatures of cerium hydride at either ambient or high pressures are lacking. In this study, we employ high-pressure diamond anvil cell (DAC) synchrotron XRD measurements, together with MD and DFT simulations, to provide better constraints on the cerium hydride EOS. Additionally, we report the high-pressure melt line of cerium hydride based on laser-heated DAC XRD experiments and MD simulations.

II. METHODS

A. High-pressure x-ray diffraction

High-pressure synchrotron XRD measurements were performed on three different samples of cerium hydride in DACs. All samples were prepared and loaded into the DACs in a

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positive pressure glovebox with an Ar atmosphere (< 0.1 ppm O_2 and < 0.5 ppm H_2O). Ambient-temperature high-pressure DAC measurements were performed on two of the three sample preparations. First, a commercially purchased (Stanford Advanced Materials) CeH_{2-3} sample was loaded in a Ne pressure transmitting medium (PTM) at a pressure of ~ 1 GPa to ensure a hydrostatic sample environment. In the second case, a flake of Ce metal was scraped from a high-purity ingot ($\geq 99.9\%$ metals basis) using a diamond scribe and was loaded in H_2 gas at a pressure of ~ 1 GPa. In this preparation, the H_2 served as both the PTM and as a reactant to synthesize cerium hydride. For the high-temperature laser-heated experiments, flakes of Ce metal were scraped from an ingot and loaded in the DAC with ammonia borane (NH_3BH_3) powder, which served as both the PTM and hydrogen source. Trace amounts of CeO_2 were present in the metallic Ce sample before laser heating, but CeO_2 peaks could not be resolved after heating the sample to form the hydride phase. In at least one case, the Ce precursor began to react with the NH_3BH_3 prior to laser heating forming small amounts of hydride (Figure S1 [22]).

In addition to the sample and hydrogen source, $10\text{-}\mu\text{m}$ Au spheres were loaded into each DAC as an internal pressure standard. The samples in NH_3BH_3 were laser heated at the closing pressure ($\sim 0.5\text{--}1$ GPa) to stimulate the reaction between Ce and NH_3BH_3 into cerium hydride. For this synthesis step, the samples were heated by pulsing them $10\text{--}50$ ms at a time with a 1064-nm , 100-W ytterbium fiber infrared laser [23]. Laser power was increased in increments of 250 mW until the laser coupled with the sample and the XRD patterns showed full conversion from Ce metal to cerium hydride. An attempt was made to laser heat the commercially purchased cerium hydride sample prepared in a KCl PTM, but the sample immediately reacted with the KCl upon heating. Depending on the preparation of the DACs, either ruby fluorescence [24] or Au lattice volumes [25] were used to determine the sample pressure.

XRD experiments were performed during two beam-times at HPCAT (High Pressure Collaborative Access Team) Beamline 16 ID-B of the Advanced Photon Source, Argonne National Laboratory, using beam energies of 25.5 and 29.2 keV, respectively. The x-ray beam was focused to a nominal spot size of $1\text{ }\mu\text{m}^2$ full width at half of maximum intensity and two-dimensional diffraction patterns were recorded on a Dectris Pilatus 2M detector. For all experiments, pressure in the DACs was controlled remotely using a gas membrane [26]. XRD images were integrated into one-dimensional patterns using DIOPTAS [27]. Cerium hydride (eight peaks) and Au (four to seven peaks) peak positions were fit in GSAS-II [28] and used to calculate unit cell volumes. Unit cell volumes of Au were used to calculate sample pressures based on a previously reported EOS [25]. Pressure-volume data, along with their associated uncertainties, were fit to the Vinet EOS [29] to determine the zero-pressure volume, isothermal bulk modulus, and pressure derivative of the bulk modulus (V_0 , $B_{0,T}$, $B'_{0,T}$).

B. High-pressure laser heating

Double-sided square pulse laser heating was used to heat the sample in $10\text{--}50$ ms bursts. The laser spot size was focused

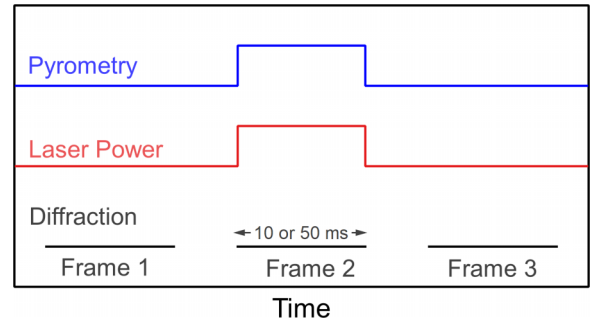


FIG. 1. Schematic for laser square pulse heating and XRD measurements. Temperature is determined pyrometrically during Frame 2. XRD acquisition, heating pulse, and pyrometry measurement times were either 10 or 50 ms. The time between XRD frames was 20 or 100 ms.

to an area of roughly $20\text{ }\mu\text{m}^2$ at the sample position. Three sequential XRD patterns were collected: (1) immediately before heating, (2) during heating, and (3) immediately after quenching the sample as depicted schematically in Fig. 1. The exposure times of the three diffraction patterns and the duration of the heating varied between experiments and were all 10 or 50 ms. Beamline-provided software was used to determine temperature from the spectral radiance of the light emitted from the sample during heating. Light emitted from the sample is assumed to be proportional to the spectral radiance of a blackbody, modified by a transfer function that includes system optics, detector response, and temperature- and wavelength-dependent material emissivity. The measured spectrum is fit to a scaled version of Planck's Law,

$$I(\lambda, T) = A \frac{c_1}{\lambda^5 [\exp(\frac{c_2}{\lambda T}) - 1]}, \quad (1)$$

where λ is the wavelength (m). T (temperature in K) and A are fit parameters (A has units of m^3/W and incorporates emissivity, collection efficiency, detector response, and geometric factors). Sample emissivity is assumed to be constant over the fitted spectral range and c_1 and c_2 are radiation constants ($c_1 = 3.7418 \times 10^{-16} \text{ W m}^2$ and $c_2 = 0.014388 \text{ m K}$) [23]. At each pressure, laser power was increased in 250 mW increments until the onset of melt was identified as described below.

C. Density functional theory and molecular dynamics

DFT calculations were performed using the HSE06 functional with collinear spin polarization in VASP [30], as have been used for Ce metal [31]. Calculations were performed on a crystallographic unit cell with an $8 \times 8 \times 8$ k -point mesh and 350 eV plane-wave cutoff. Gaussian smearing was used with a smearing width of 0.03 eV. The convergence criterion was set to a cutoff of 10^{-7} eV. Initial magnetic ordering was set to be ferromagnetic and no phonon relaxation occurred. MD simulations were run using a previously developed machine learning interatomic potential (MLIAP) [32] using the hierarchically interacting particle neural network (HIPNN) model [33]. Simulations were run with the LAMMPS package [34] under isothermal-isobaric conditions (NVT) at 300 K with pressure as the control variable. A 0.2 fs time step and

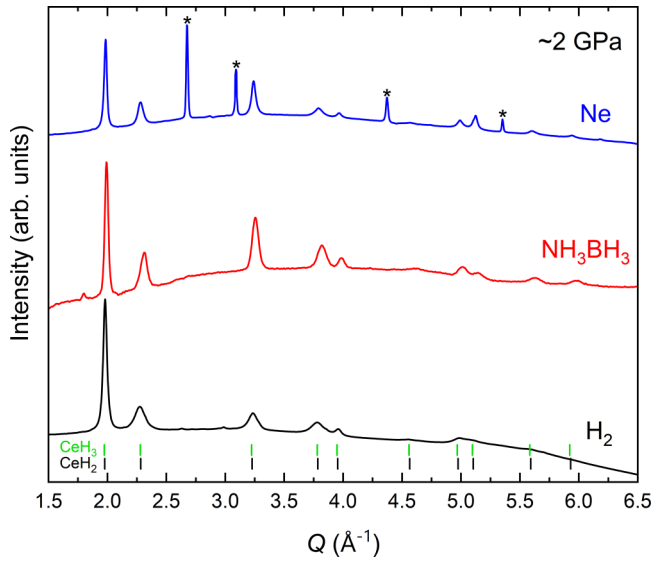


FIG. 2. Comparison of XRD patterns collected from cerium hydride samples described in this study at ~ 2 GPa. The blue curve was measured on a commercially purchased sample (asterisks indicate peaks from an Au internal pressure standard). The red curve was measured on a sample of cerium hydride synthesized by laser heating Ce metal in the presence of NH_3BH_3 in the DAC. The black curve was measured on a sample of cerium hydride synthesized by loading a sample of Ce metal with H_2 in the DAC (no heating). Green and black ticks show predicted peak positions for end-member CeH_3 and CeH_2 compositions at ~ 2 GPa, respectively, using EOS parameters from Ref. [14].

$10 \times 10 \times 10$ replication of the unit cell were used in all cases.

III. RESULTS AND DISCUSSION

A. High-pressure equation of state

A comparison of low-pressure, ambient temperature, XRD patterns collected from samples prepared using the three methods described above used is shown Fig. 2. The lattice volumes for each sample are $V_0^{\text{Ne}} = 165.47(26)$, $V_0^{\text{H}_2} = 165.16(43)$, and $V_0^{\text{NH}_3\text{BH}_3} = 162.13(60)$ $\text{\AA}^3$. The commercially purchased $\text{CeH}_{2.3}$ powder loaded in Ne has sharp, well-defined peaks. The patterns measured on samples loaded in NH_3BH_3 and H_2 have considerably broader peaks. Peak broadening associated with these samples is likely a result of microstrain. For the sample loaded in NH_3BH_3 , the microstrain is likely due to local nonhydrostatic conditions arising from the solid pressure medium. The sample loaded in H_2 was not annealed after the DAC was initially sealed, and the pattern shown in Fig. 2 reflects preserved defects associated with diffusion of H_2 into the Ce crystal lattice. Both CeH_2 and CeH_3 have been reported as the equilibrium phase when Ce metal is contained in an overpressure of H_2 [10,35]. The peak locations plotted in Fig. 2 show the structural similarities between CeH_2 and CeH_3 , and emphasize the difficulty in using powder diffraction patterns alone to differentiate between the two isostructural end-member compositions. However, fitted

zero-pressure volumes produced from our EOS fits below suggest that our samples are hydrogen rich, with $\text{H}/\text{Ce} > 2$.

High-pressure XRD measurements from the DAC experiments, along with data from the literature, are shown in Fig. 3. Noble gas PTMs have been shown to sustain pressure gradients < 0.2 GPa at the conditions of this study [36]. While NH_3BH_3 is commonly used as a hydrogen source and PTM in high-pressure metal hydride experiments, it has not been evaluated as a PTM. However, similarly soft materials such as NaCl and KCl have been shown to sustain pressure gradients on the order of ~ 0.5 GPa in the DAC at pressures up to 20 GPa [37]. Uncertainties in unit cell volumes were propagated into pressure via partial derivatives of the Vinet equation of state. That is,

$$\delta P = \sqrt{\left(\frac{\partial P}{\partial B_{0,T}} \delta B_{0,T}\right)^2 + \left(\frac{\partial P}{\partial B'_{0,T}} \delta B'_{0,T}\right)^2 + \left(\frac{\partial P}{\partial V} \delta V\right)^2}. \quad (2)$$

The resulting error bars are smaller than the plotted symbols in Fig. 3. Our data show considerably less scatter in pressure-volume space than previous measurements. Previous experimental studies [4–7] were primarily concerned with structural refinements and electrical conductivity measurements, rather than precise EOS determination. Consequently, data coverage from those experiments in pressure-volume space is comparatively sparse. Furthermore, with the exception of Ref. [6], these experiments used the diamond anvil Raman gauge for pressure determination [38] which is unsuitable for estimating pressure gradients across the sample chamber.

The datasets measured in Ne and H_2 pressure media are consistent with each other and show no indication of a first-order phase transition up to 20 GPa. Our data are well-described by a Vinet EOS, with fit parameters reported in Table I. EOS were fit to data separately from each experiment, as well as to the combined data. Fit zero-pressure volumes range from 168.21(21) to 169.56(41) $\text{\AA}^3$ and the fit bulk moduli range from 76.7(51) to 85.8(23) GPa. Compared to the bulk modulus of 21 GPa for γ -Ce [39], 26 GPa for α -Ce [39], 34 GPa for α'' -Ce, and 66 GPa for ϵ -Ce [40], these values indicate that cerium hydride is substantially stiffer than all elemental phases of Ce present at pressures for which the cerium hydride P - V data were collected. The trend of enhanced incompressibility upon hydriding is common among many metals, including those in the lanthanide series where the effect has been observed in lanthanum [41,42], praseodymium [43,44], neodymium [45,46], and dysprosium [47,48].

Results from DFT and MD simulations are shown in Figs. 3(b) and 3(c) alongside the experimental DAC data. The DFT simulations were performed at 0 K and the MD simulations were performed at 300 K. Vinet EOS were fit to each simulated dataset and are reported in Table I. The DFT calculations predict a strong compositional effect on volume at low pressures, with the two end-member compositions predicted to have a 6% volume difference at ambient pressure. The compression curves generated from the MD simulations show a moderate compositional dependence. While CeH_3 is initially denser than CeH_2 in these simulations, CeH_2 is predicted to be more compressible and the two curves intersect at

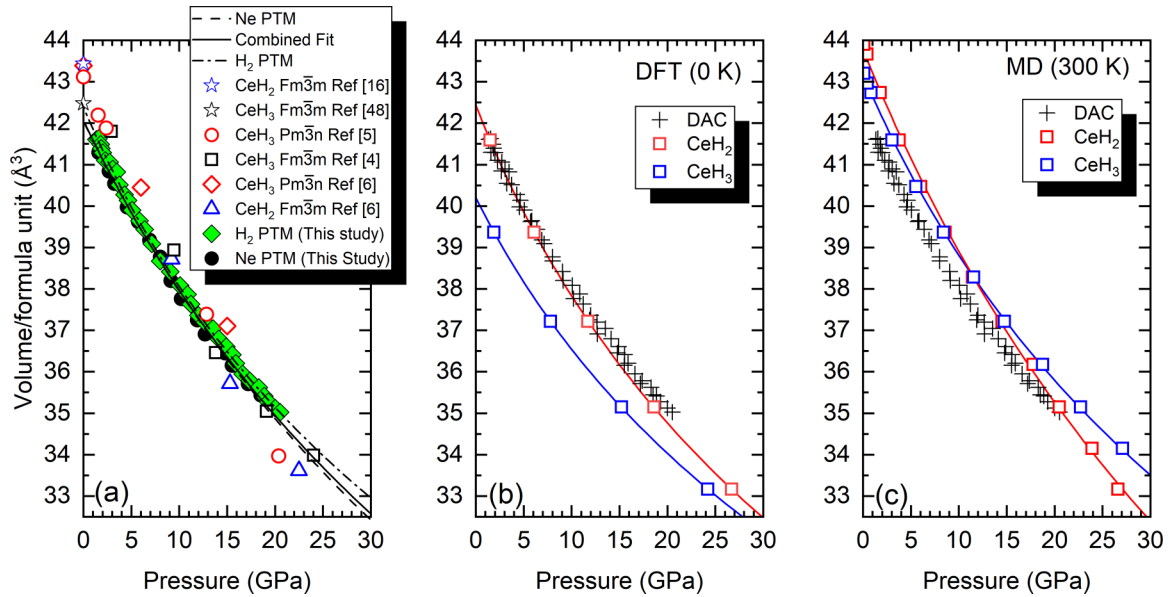


FIG. 3. (a) High-pressure ambient-temperature EOS measurements of cerium hydrides from the literature and this study. Data from this study are plotted as solid symbols and literature values [4–6,16,49] are plotted as open symbols. The sample compressed in Ne was commercially purchased and the sample compressed in H₂ was synthesized in the DAC. Vinet EOS fits are plotted as solid and broken black lines. Results from (b) DFT and (c) MD simulations for CeH₂ and CeH₃ at 0 and 300 K, respectively. Experimental data are plotted as “+” symbols for comparison. Fit parameters for each curve are reported in Table I.

~12 GPa, with CeH₂ remaining denser upon further compression. This density crossover reflects structural differences between the two end-member compositions: octahedral sites are fully vacant in CeH₂ but occupied in CeH₃. So, while increased compressibility is predicted for both compositions upon heating (i.e., $dB_0/dT < 0$), the effect is tempered in CeH₃ by the occupied octahedral sites.

B. High-pressure melting experiments and theoretical predictions

Uncertainties in temperature and pressure are undoubtedly correlated in laser-heated DAC (LHDAC) experiments. Due to experimental limitations and a lack of thermal expansion data with respect to our choice of PTM and sample, our pressures are reported at ambient temperature and uncorrected for the effects of the thermal pressure, $(\partial P/\partial T)_V$. Hence, our reported pressures are likely underestimated. It has been previously shown that the thermal pressure is strongly localized around the laser spot, has magnitude that scales as $\alpha B_T \Delta T$, and decays at a rate of 0.5–1.0 GPa/10 μm from the hot spot [50]. As an upper bound for estimating the thermal pressure, we assume a sample chamber filled only with CeH_{2–3} with $\alpha \sim 10^{-5} \text{ K}^{-1}$, $B_T \sim 10^2 \text{ GPa}$, $\Delta T \sim 10^3 \text{ K}$, and find that uncertainties in pressure due to thermal effects

would be on the order of ~1 GPa. Since the sample chambers in these experiments were mostly filled with (partially dehydrogenated) NH₃BH₃ and included only small flakes of CeH_{2–3}, we anticipate uncertainties due to thermal pressure to be less than the estimated +1 GPa upper limit. For the laser-heated DAC measurements, temperatures and associated uncertainties for individual measurements were calculated following the method of Ref. [51]. This method incorporates differences between upstream (T_{up}) and downstream (T_{down}) temperatures, wavelength-dependent emissivity, system response calibration, Planck fit errors, and effects associated with radial temperature gradients:

$$T = \frac{T_{\text{up}} + T_{\text{down}}}{2} \pm \left[\frac{|T_{\text{up}} - T_{\text{down}}|}{2} + 0.015 \left(\frac{T_{\text{up}} + T_{\text{down}}}{2} \right) + 50 \right]. \quad (3)$$

T_{up} and T_{down} refer to pyrometric measurements from the two sides of the sample determined using Eq. (1). Determination of the onset of melting was informed by the observation of qualitative changes in two dimensions (2D) and integrated diffraction patterns [i.e., weakening of the Bragg ring and integrated peak intensity, as shown in Figs. 4(a) and 4(b)].

TABLE I. Vinet EOS fit parameters for cerium hydride DAC experiments and simulations. Experimental fit parameters reflect uncertainties in both pressure and volume. Covariance matrices for the fits are reported in the Supplemental Material [22].

	Ne PTM	H ₂ PTM	Combined	CeH ₂ 0 K (DFT)	CeH ₂ 300 K (MD)	CeH ₃ 0 K (DFT)	CeH ₃ (300 K) MD
V_0 (Å ³)	168.21(21)	169.56(41)	168.28(13)	169.74(15)	175.11(48)	160.82(13)	172.71(19)
$B_{0,T}$ (GPa)	87.2(39)	76.7(51)	85.8(23)	71.6(12)	76.2(39)	86.9(12)	78.9(17)
$B'_{0,T}$	2.2(6)	3.6(6)	2.5(3)	3.5(1)	1.8(4)	4.0(1)	3.3(2)

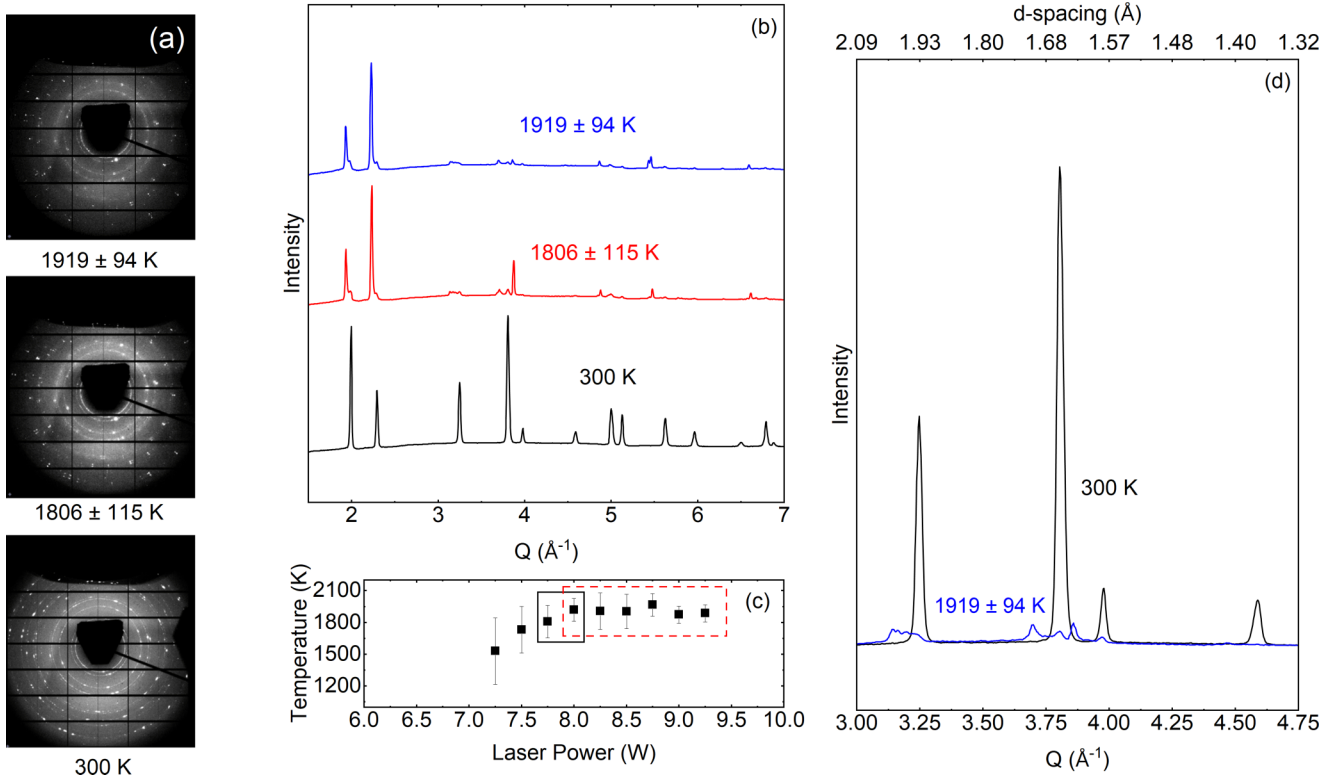


FIG. 4. Two-dimensional diffraction images collected at 3.7 GPa and 300, 1806 (just before melting), and 1919 K (first appearance of melt) are shown in (a). Full resolution images of (a) are included in the Supplemental Material [22]. Corresponding integrated one-dimensional patterns are shown in (b). Collection times are the same for all patterns. Laser power-temperature relation at 3.7 GPa is shown in (c). The plateau used for melt temperature determination is bounded by the dashed red box in (c) and integrated patterns for the boxed points in (c) are shown in (b). (d) shows movement of four select peaks and reduction in peak intensity due to thermal expansion and the onset of melting. In addition to the features associated with melt, low intensity peaks of the cold material are visible due to the x-ray beam tails sampling material outside of the molten region.

Melting was inferred to initiate at the onset of the temperature-laser power plateau [red boxed points in Fig. 4(c)]. Diffuse scattering from the liquid was not observed in the diffraction patterns. This was likely the result of short acquisition times and the tails of the x-ray beam probing crystalline material outside of the melt pool. Reported melting temperatures and uncertainties correspond to the weighted mean and standard deviation of points in the temperature-laser power plateau [i.e., red dashed box in Fig. 4(c)]. While the selection of points to include in this plateau is subjective, the inclusion of one more or one fewer point had only a slight effect on the determined temperature. For example, adding or removing one point from the red dashed box region in Fig. 5(c) changed the determined melt temperature, T_m , by <10 K. For a single temperature measurement t_i with uncertainty σ_i in a plateau containing N measurements, the reported melting temperature is determined by

$$T_m = \frac{\sum_{i=1}^N t_i / \sigma_i^2}{\sum_{i=1}^N 1 / \sigma_i^2} \pm \sqrt{\frac{N \sum_{i=1}^N \frac{(t_i - T_m)^2}{\sigma_i^2}}{(N-1) \sum_{i=1}^N \frac{1}{\sigma_i^2}}}. \quad (4)$$

The high-pressure melting curves of cerium hydride and Ce metal (reproduced from Ref. [52]) are shown in Fig. 5(a). A linear fit to the DAC data yields an ambient-pressure melting temperature of $T_0 = 1737(75)$ K and a slope of $(\frac{\partial T}{\partial P})_{\Delta G=0} =$

27(15) K/GPa, where P and G are the pressure and Gibbs free energy, respectively. Our fit zero-pressure melting temperature is approximately 5% higher than the maximum temperature reported in shock ejecta studies [3]. The melting curve of CeH₂, generated from MD simulations, is also shown in Fig. 5(a). The trend is nonlinear up to 0.5 GPa. Above 0.5 GPa, the trend becomes linear with fit parameters $(\frac{\partial T}{\partial P})_{\Delta G=0} = 16(0)$ K/GPa and $T_0 = 1671(0)$ K. While the melting curves of cerium hydride and cerium metal have similar slopes above ~ 4 GPa, cerium hydride consistently melts at higher temperatures than cerium metal. This systematic difference in melting temperature reflects the stronger ionic bonds in cerium hydride compared to the metallic bonding environment in Ce. In contrast to Ce metal, the slope of the cerium hydride melting curve is constant and positive over the range of pressures measured in this study.

Figure 5(a) also shows results from a model based on Lindemann melting theory [53,54], which has been modified to allow for the prediction of melting temperatures at high pressures by incorporating the shear modulus and its pressure dependence along the melt curve at high pressures (Ref. [49] and Supplemental Material [22]). In this framework, the melting temperature is

$$T_m = \lambda^2 \left(\frac{\pi}{3} \right)^{\frac{4}{3}} \left(\frac{a_{nn}}{a} \right)^2 \frac{M(\text{Ce})}{M(\text{CeD}_2)} \frac{a^3 \mu(\rho_m, T_m)}{k_B}, \quad (5)$$

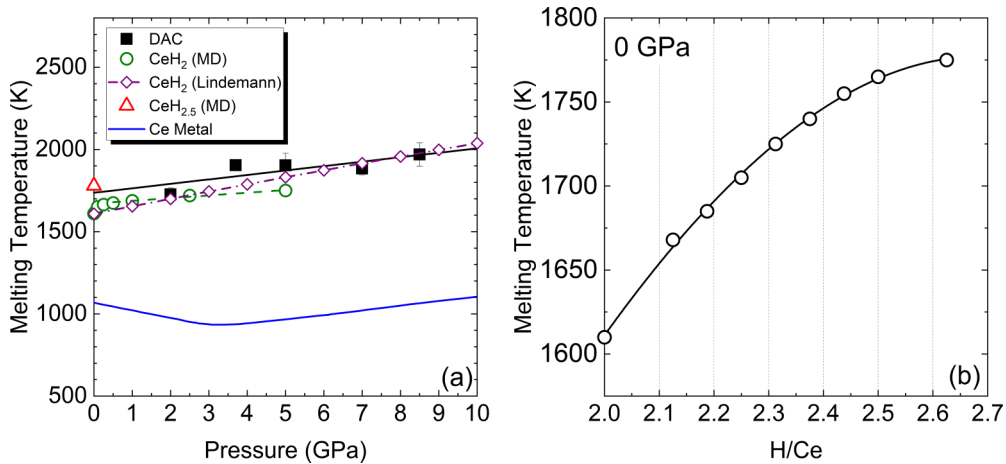


FIG. 5. Melting curves of Ce metal (reproduced from Ref. [52]) and cerium hydride (this study) are shown in (a). Compositional dependence of the zero-pressure melting temperature of cerium hydride from MD simulations is shown in (b), reproduced from Ref. [32]. The cubic fit in (b) is a guide for the eye.

where λ , the Lindemann ratio, is the root-mean-square Ce atom displacement divided by the Ce-H nearest-neighbor separation a_{nn} , and a is the cubic unit cell lattice constant. M is the atomic mass, μ is the shear modulus, ρ is the mass density, and k_B is the Boltzmann constant. Equation (5) can be rewritten in terms of the unit cell volume as

$$\frac{T_{m,P}}{T_{m,P=0}} = \frac{V}{V_0} \frac{\mu_{T_{m,P}}}{\mu_{T_{m,P=0}}}, \quad (6)$$

where the pressure dependence of the shear modulus along the melting curve is approximated as

$$\mu_P = \mu_{P=0} \left(1 + \mu' \frac{P}{\mu_{P=0}} \right). \quad (7)$$

Since the pressure dependence of the shear modulus of CeH_2 is not known, melting curves were calculated for a range of values of μ' from 0.5 to 2.0 for $\mu_0 = 37.16$ GPa [55]. We note here that a value of $\mu' = 1.5$, close to the value of $\mu' = 1.3$ reported by Ref. [56] for Ce metal, yields general agreement with the DAC data. With $T_{m,P=0} = 1609$ K, this modified Lindemann method yields a slope of $(\frac{\partial T}{\partial P})_{\Delta G=0} = 44$ K/GPa.

The compositional dependence of the melting temperature of cerium hydride was modeled in MD simulations, and is plotted in Fig. 5(b) (reproduced from Ref. [32]). The melting temperature increases nonlinearly from 1610 to 1775 K over the compositional range $\text{H/Ce} = 2.0$ – 2.6 . The melting curve is particularly sensitive to compositional changes for low values of H/Ce . Indeed, a change in composition from CeH_2 to $\text{CeH}_{2.1}$ corresponds to an increase in melting temperature of greater than 460 K, while a change in composition from $\text{CeH}_{2.5}$ to $\text{CeH}_{2.63}$ corresponds to an increase of 80 K. In agreement with the high-pressure EOS data, the fit zero-pressure melt temperature of 1737 K determined from the DAC data and the compositional melting relation shown in Fig. 5(b) indicate a hydrogen-rich sample.

IV. CONCLUSIONS

Using experimental techniques (synchrotron XRD of samples compressed in DACs) and calculational methods (DFT and MD), we provide improved constraints on the bulk modulus of cerium hydride, bounding the bulk modulus to the range 76.2(39)–87.2(39) GPa at 300 K. DFT and MD simulations indicate an increase in both compressibility and melting temperature with increasing hydrogen content, but due to the structural similarities of the CeH_2 and CeH_3 phases and uncertainties in pyrometric temperature measurements, this compositional dependence is likely too subtle to be distinguished in the DAC.

Data from laser-heated DAC experiments indicate the melting temperature of cerium hydride increases linearly up to a pressure of at least 10 GPa, with $T_{m,0} = 1737(75)$ K and $(\frac{\partial T}{\partial P})_{\Delta G=0} = 27(15)$ K/GPa. MD simulations of CeH_2 predict an initial nonlinear dependence of melting temperature on pressure up to 0.5 GPa, after which the trend becomes linear with slope $(\frac{\partial T}{\partial P})_{\Delta G=0} = 16(0)$ K/GPa. A simple, modified Lindemann melting model for CeH_2 yields a slope of $(\frac{\partial T}{\partial P})_{\Delta G=0} = 44$ K/GPa.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

- [1] A. Rossi, Crystal structure of lanthanum, cerium and praseodymium hydrides, *Nature (London)* **133**, 174 (1934).
- [2] W. T. Buttler, R. K. Schulze, J. J. Charonko, J. C. Cooley, J. E. Hammerberg, J. D. Schwarzkopf, D. G. Sheppard, J. J. Goett III, M. Grover, B. M. La Lone, S. K. Lamoreaux, R. Manzanares, J. I. Martinez, J. D. Regele, M. M. Schauer, D. W. Schmidt, G. D. Stevens, W. D. Turley, and R. J. Valencia, Understanding the transport and break up of reactive ejecta, *Physica D* **415**, 132787 (2021).
- [3] T. M. Hartsfield *et al.*, The temperatures of ejecta transporting in vacuum and gases, *J. Appl. Phys.* **131**, 195104 (2022).
- [4] X. Li *et al.*, Polyhydride CeH₉ with an atomic-like hydrogen clathrate structure, *Nat. Commun.* **10**, 3461 (2019).
- [5] X. Li, X. Huang, W. Chen, D. Zhou, H. Xie, Q. Zhuang, D. Duan, and T. Cui, New cage-like cerium trihydride stabilized at ambient conditions, *CCS Chem* **4**, 825 (2022).
- [6] N. P. Salke *et al.*, Synthesis of clathrate cerium superhydride CeH₉ at 80–100 GPa with atomic hydrogen sublattice, *Nat. Commun.* **10**, 4453 (2019).
- [7] W. Chen, D. V. Semenov, X. Huang, H. Shu, X. Li, D. Duan, T. Cui, and A. R. Oganov, High-temperature superconducting phases in cerium superhydride with a T_c up to 115 K below a pressure of 1 megabar, *Phys. Rev. Lett.* **127**, 117001 (2021).
- [8] P. Vorderwisch, S. Hautecler, and B. Dorner, Phonon dispersion relations in CeD_{2.72}, *Solid State Commun.* **34**, 853 (1980).
- [9] D. Avisar and T. Livneh, Raman scattering by phonons and crystal-field excitations in cerium hydrides, *J. Alloys Compd.* **494**, 11 (2010).
- [10] T. Livneh and D. Avisar, Raman scattering from cerium hydride growth centers overlaid by hydrogen-incorporated oxide, *J. Phys. Chem. C* **124**, 28018 (2020).
- [11] A. Fujimori, M. Ishii, and N. Tsuda, Infrared and raman spectra of non-stoichiometric cerium hydrides, *Phys. Status Solidi B* **99**, 673 (1980).
- [12] X. Ye, H. Li, and S. Li, New insights into phase separation of cerium hydrides under pressure, *ACS Omega* **7**, 15681 (2022).
- [13] T. Gürel and R. Eryiğit, Volume dependent vibrational properties of cerium hydrides from first principles, *J. Alloys Compd.* **477**, 478 (2009).
- [14] G. S. Priyanga, R. Rajeswarapalanichamy, and K. Iyakutti, First principles study of structural, electronic, elastic and magnetic properties of cerium and praseodymium hydrogen system REH_x (RE: Ce, Pr and $x = 2, 3$), *J. Rare Earths* **33**, 289 (2015).
- [15] J. A. Bjorgaard, J. Hammerberg, and D. Sheppard, Density functional theory study of cerium deuterides, *AIP Conf. Proc.* **1979**, 080002 (2018).
- [16] W. L. Korst and J. C. Warf, Rare earth-hydrogen systems. i. structural and thermodynamic properties, *Inorg. Chem.* **5**, 1719 (1966).
- [17] G. G. Libowitz, J. G. Pack, and W. P. Binnie, Temperature-dependent electronic transition in cerium hydride, *Phys. Rev. B* **6**, 4540 (1972).
- [18] E. Boroch and E. Kaldis, Phase transitions in the CeH₂-CeH₃ system, *Inorg. Chim. Acta* **140**, 89 (1987).
- [19] M. Tellefsen, E. Kaldis, and E. Jilek, The phase diagram of the Ce-H₂ system and the CeH₂-CeH₃ solid solutions, *J. Less Common Met.* **110**, 107 (1985).
- [20] E. Kaldis, E. Boroch, and M. Tellefsen, Phase relationships and phase transitions in the homogeneity range of pure CeH₂-CeH₃, *J. Less-Common Met.* **129**, 57 (1987).
- [21] E. Boroch and E. Kaldis, Progress in the T-X phase diagram of the solid solution CeH₂-CeH₃, *Z. Phys. Chem.* **163**, 117 (1989).
- [22] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/gglf-wx5w> for an overview of Lindemann melting theory and calculations used for the cerium hydride melting curve, a comparison of XRD patterns of Ce loaded in ammonia borane and cerium hydride (i.e., before and after laser heating), a stress strain plot for the high pressure EOS fit, and some high resolution XRD images.
- [23] Y. Meng, R. Hrubiak, E. Rod, R. Boehler, and G. Shen, New developments in laser-heated diamond anvil cell with *in situ* synchrotron x-ray diffraction at High Pressure Collaborative Access Team, *Rev. Sci. Instrum.* **86**, 072201 (2015).
- [24] G. Shen *et al.*, Toward an international practical pressure scale: A proposal for an IPPS ruby gauge (IPPS-Ruby2020), *High Press. Res.* **40**, 299 (2020).
- [25] A. Dewaele, P. Loubeyre, and M. Mezouar, Equations of state of six metals above 94 GPa, *Phys. Rev. B* **70**, 094112 (2004).
- [26] S. V. Sinogeikin, J. S. Smith, E. Rod, C. Lin, C. Kenney-Benson, and G. Shen, Online remote control systems for static and dynamic compression and decompression using diamond anvil cells, *Rev. Sci. Instrum.* **86**, 072209 (2015).
- [27] C. Prescher and V. B. Prakapenka, *DIOPTAS*: A program for reduction of two-dimensional X-ray diffraction data and data exploration, *High Pressure Res.* **35**, 223 (2015).
- [28] B. H. Toby and R. B. Von Dreele, GSAS-II: The genesis of a modern open-source all purpose crystallography software package, *J. Appl. Crystallogr.* **46**, 544 (2013).
- [29] P. Vinet, J. Ferrante, J. H. Rose, and J. R. Smith, Compressibility of solids, *J. Geophys. Res.* **92**, 9319 (1987).
- [30] G. Kresse and J. Furthmüller, Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [31] B. W. Hamilton, A. R. Muñoz, T. E. Jones, and B. T. Nebgen, Mott vs Kondo: Influence of various density functional based methods on the Ce isostructural phase transition mechanism, *J. Appl. Phys.* **139**, 055101 (2026).
- [32] B. W. Hamilton, T. E. Jones, T. C. Germann, and B. T. Nebgen, Stoichiometry dependent properties of cerium hydride: An active learning developed interatomic potential study, *Phys. Rev. Mater.* **10**, 073603 (2026).
- [33] N. Lubbers, J. S. Smith, and K. Barros, Hierarchical modeling of molecular energies using a deep neural network, *J. Chem. Phys.* **148**, 241715 (2018).

- [34] A. P. Thompson *et al.*, LAMMPS - a flexible simulation tool for particle-based materials modeling at the atomic, meso, and continuum scales, *Comput. Phys. Commun.* **271**, 108171 (2022).
- [35] Y. Wang, Y. Ge, S. Yu, S. Ma, X. Huang, P. Zhu, S. Jiang, X. Li, and T. Cui, Synthesizing cerium hydrides with high hydrogen saturation under high temperature and pressure conditions, *Solid State Commun.* **404**, 116084 (2025).
- [36] S. Klotz, J.-C. Chervin, P. Munsch, and G. Le Marchand, Hydrostatic limits of 11 pressure transmitting media, *J. Phys. D Appl. Phys.* **42**, 075413 (2009).
- [37] I. Uts, K. Glazyrin, and K. K. M. Lee, Effect of laser annealing of pressure gradients in a diamond-anvil cell using common solid pressure media, *Rev. Sci. Instrum.* **84**, 103904 (2013).
- [38] Y. Akahama and H. Kawamura, Pressure calibration of diamond anvil Raman gauge to 310 GPa, *J. Appl. Phys.* **100**, 043516 (2006).
- [39] J. S. Olsen, L. Gerward, U. Benedict, and J. P. Itié, The crystal structure and the equation of state of cerium metal in the pressure range 0–46 GPa, *Physica B+C* **133**, 129 (1985).
- [40] C. Ma, Z.-Y. Dou, H.-Y. Zhu, G.-Y. Fu, X. Tan, B. Bai, P.-C. Zhang, and Q.-L. Cui, Structure phase transformation and equation of state of cerium metal under pressures up to 51 GPa, *Chin. Phys. B* **25**, 046401 (2016).
- [41] W. Chen, D. V. Semenok, I. A. Troyan, A. G. Ivanova, X. Huang, A. R. Oganov, and T. Cui, Superconductivity and equation of state of lanthanum at megabar pressures, *Phys. Rev. B* **102**, 134510 (2020).
- [42] A. Machida, T. Watanuki, D. Kawana, and K. Aoki, Phase separation of lanthanum hydride under high pressure, *Phys. Rev. B* **83**, 054103 (2011).
- [43] B. J. Baer, H. Cynn, V. Iota, C.-S. Yoo, and G. Shen, Phase diagram and equation of state of praseodymium at high pressures and temperatures, *Phys. Rev. B* **67**, 134115 (2003).
- [44] D. Zhou, D. V. Semenok, D. Duan, H. Xie, W. Chen, X. Huang, X. Li, B. Liu, A. R. Oganov, and T. Cui, Superconducting praseodymium superhydrides, *Sci. Adv.* **6**, eaax6849 (2020).
- [45] S. E. Finnegan, C. V. Storm, E. J. Pace, M. I. McMahon, S. G. MacLeod, E. Plekhanov, N. Bonini, and C. Weber, High-pressure structural systematics in neodymium up to 302 GPa, *Phys. Rev. B* **103**, 134117 (2021).
- [46] D. Zhou *et al.*, High-pressure synthesis of magnetic neodymium polyhydrides, *J. Am. Chem. Soc.* **142**, 2803 (2020).
- [47] D. T. Sneed, P. Söderlind, E. F. O'Bannon, H. Cynn, D. Smith, J. S. Smith, C. Park, and Zs. Jenei, High-pressure structural systematics of dysprosium metal compressed in a neon pressure medium to 182 GPa, *Phys. Rev. B* **105**, 214110 (2022).
- [48] H. Meng, T. Palasyuk, V. Drozd, and M. Tkacz, Study of phase stability and isotope effect in dysprosium trihydride at high pressure, *J. Alloys Compd.* **722**, 946 (2017).
- [49] M. E. Kost, N. T. Kuznetsov, and A. L. Shilov, New hydride phases in the systems R-Mg-H (R = Y, La, Ce), *Dokl. Chem. (Engl. Transl.)* **292**, 632 (1987).
- [50] C. E. Yen, Q. Williams, and M. Kunz, Thermal pressure in the laser-heated diamond anvil cell: A quantitative study and implications for the density versus mineralogy correlation of the mantle, *JGR Solid Earth* **125**, e2020JB020006 (2020).
- [51] J. S. Pigott, N. Velisavljevic, E. K. Moss, N. Draganic, M. K. Jacobsen, Y. Meng, R. Hrubak, and B. T. Sturtevant, Experimental melting curve of zirconium metal to 37 GPa, *J. Phys.: Condens. Matter* **32**, 355402 (2020).
- [52] A. Schiwek, F. Porsch, and W. B. Holzapfel, High temperature-high pressure structural studies of cerium, *High Pressure Res.* **22**, 407 (2002).
- [53] F. Lindemann, The calculation of molecular eigenfrequencies, *Phys. Z.* **11**, 609 (1910).
- [54] L. Burakovsky, C. W. Greeff, and D. L. Preston, Analytic model of the shear modulus at all temperatures and densities, *Phys. Rev. B* **67**, 094107 (2003).
- [55] P. Vorderwisch, S. Hautecler, and W. D. Teuchert, Phonon dispersion relations in CeD_{2.12}, *Solid State Commun.* **25**, 213 (1978).
- [56] M. Guinan and D. Steinberg, Pressure and temperature derivatives of the isotropic polycrystalline shear modulus for 65 elements, *J. Phys. Chem. Solids* **35**, 1501 (1974).
- [57] <https://doi.org/10.46936/APS-193059/60016244>.