The discovery of two-dimensional (2D) van der Waals (vdW) materials often provides interesting playgrounds to explore novel phenomena. One of the missing components in 2D vdW materials is the intrinsic heavy-fermion systems, which can provide an additional degree of freedom to study quantum critical point (QCP), unconventional superconductivity, and emergent phenomena in vdW heterostructures. Here, we investigate 2D vdW heavy-fermion candidates through the database of experimentally known compounds based on dynamical mean-field theory calculation combined with density functional theory (DFT+DMFT). We have found that the Kondo resonance state of CeSiI does not change upon exfoliation and can be easily controlled by strain and surface doping. Our result indicates that CeSiI is an ideal 2D vdW heavy-fermion material and the quantum critical point can be identified by external perturbations.
RESULTS

Screening from the database

We first briefly investigate the electronic structures for 32 layered lanthanide materials proposed to be exfoliable. Not only localized f electrons but also itinerant conduction electrons are essential to forming a heavy-fermion state. Thus, one can easily get an important clue to identify heavy-fermion materials by simply checking the conduction bands at the Fermi level (\(E_F\)). Therefore, we perform DFT open-core calculations, which consider f electrons as core electrons, to examine the existence of non-f conduction bands at \(E_F\).

Figure 1 shows the density of states (DOS) at \(E_F\) for 32 layered lanthanide materials obtained from DFT calculation within the open-core approach. Open symbols indicate La (\(f^0\)) and Lu (\(f^{14}\)) compounds. Since they have empty or fully occupied 4f shells, the lattice Kondo effect cannot be expected. On the other hand, the compounds denoted by square symbols also cannot be heavy-fermion compounds since they exhibit an insulating nature, indicating that there are no itinerant conduction bands to screen localized 4f electrons. Finally, we are left with only four compounds (CeSiI, ErHCl, TmI2, and YbI2) satisfying the minimal necessary conditions. The four remaining candidates can be again divided into two categories. The conduction bands of CeSiI at \(E_F\) are mainly coming from the ligand atoms, while those of other compounds are coming from solely lanthanide 5d orbitals.

For comparison, \(\alpha\)-Ce is a typical heavy-fermion system whose 4f electron is screened by its own 5d orbitals. In contrast, \(\gamma\)-Ce, which has an isostructure with a larger volume, shows weak 4f-5d hybridization yielding a small Kondo resonance peak\(^{29,30}\). Since ErHCl, TmI\(_2\), and YbI\(_2\) have ligand atoms between their lanthanide atoms, the distance between the lanthanide atoms of those compounds is even greater than that of \(\gamma\)-Ce (3.65 Å). Therefore, 4f electrons are hardly screened by neighboring lanthanide 5d orbitals or the Kondo cloud at each lanthanide site hardly sees each other to form the coherent heavy-fermion state.

Although CeI\(_2\) is not classified into exfoliable compounds from the previous study\(^{28}\), it has the same structure as TmI\(_2\) and YbI\(_2\). CeI\(_2\) has a magnetically ordered ground state and the previous photoemission spectroscopy (PES) study showed that the \(f^0\) peak is almost absent at \(E_F\), indicating 4f-5d hybridization in CeI\(_2\) is much smaller than \(\alpha\) and \(\gamma\)-Ce cases\(^{30}\). Our DFT+DMFT calculation on CeI\(_2\) also verifies the absence of a Kondo peak near \(E_F\) (see Supplementary Fig. 1). Considering these aspects, we conclude CeSiI is the most promising candidate for 2D vdW heavy-fermion systems among 32 exfoliable lanthanide compounds. Very recently, it was reported that bulk CeSiI has a magnetically ordered ground state\(^{31}\). However, it can be sensitively tuned by external perturbations, which will be discussed in later sections.

Structural and electronic properties of CeSiI

The CeSiI vdW layer consists of a silicene layer, surrounding triangular Ce cage (orange dashed line), and outer iodine layers. The vdW gap can be defined as the iodine to iodine as depicted in Fig. 2a. The electronic structures calculated by DFT within the open-core approach are presented in Fig. 2b in terms of DOS (right) and band dispersion (left). Analysis of DOS and band dispersion leads to the following observation: (1) The iodine \(p\) state occurs primarily between \(-5\) and \(-2\) eV indicating that the iodine \(p\) state is fully occupied, and their contribution at \(E_F\) is negligible. (2) The band dispersion near \(E_F\) resembles that of pure silicene compound while the Dirac point at \(K\) point is moved to \(-2\) eV in contrast to pure silicene, where the Dirac point is located at \(E_F\). This indicates there is charge transfer from the Ce atom to the silicene layer. (3) Therefore, the bands crossing \(E_F\) should be coming from Si \(p_x\) and \(p_y\) antibonding state. However, the contribution from Si at \(E_F\) is quite small compared to the total DOS as shown in the right panel of Fig. 2b.

The most of contribution at \(E_F\) is originated from the interstitial region. Figure 2c shows the calculated charge density distribution near \(E_F\) (energy range \(-1.5\) to \(1.5\) eV with respect to \(E_F\)) for CeSiI on (110) plane (see the red thick line in the lower left panel in Fig. 2a). The electron density is delocalized all over the Ce cage (between Ce atoms and silicene layer), not localized at specific atoms. This feature agrees well with the Zintl–Klemm feature of \([\text{Ce}^{3+}\cdot\text{Si}^-]\) as suggested by the previous study\(^{32}\). Floating electrons on the silicene layer and the interstitial region can be the ingredient of a heavy-fermion state.

To investigate the hybridization between Ce 4f and conduction electrons (\(f\rightarrow c\) hybridization) properly, DFT+DMFT calculations are performed. The calculated spectral function of bulk and monolayer CeSiI are shown in Fig. 3a at 10 K. Unlike CeI\(_2\) case, the Kondo resonance peak is clearly observed near \(E_F\) indicating substantial hybridization between Ce 4f and conduction electrons. Therefore, CeSiI is located in the vicinity of the PM heavy-fermion state. The electronic structures of bulk and monolayer CeSiI are almost identical, from which similar chemical and physical properties are expected. The temperature-dependent evolution of the Kondo resonance peak also does not change upon exfoliation as shown in the inset of Fig. 3a, b. Therefore, the CeSiI monolayer can provide an ideal platform for exploring the heavy-fermion state in the 2D monolayer limit.
hybridization under compressive strain due to the enhanced
conduction electron density in the Ce cage. Figure 4a shows the
spectral function of Ce 4f state at 10 K depending on the strain. The
Kondo resonance peak is sensitively affected by the strain.

The T-dependent 4f spectral function at \( E_F \), \( A_{4f}(\omega = 0) \) is shown in
Fig. 4b. The Kondo resonance peak is enhanced by the compressive
strain for a wide temperature range, resulting in a higher Kondo
temperature, \( T_K \). \( T_K \) can be estimated from the logarithmic \( T \) dependence behavior of \( A_{4f}(\omega = 0) = \ln(T_K/T) \) (dashed line in Fig. 4b)34,35. The estimated \( T_K \) for monolayer without strain is
120 K. \( T_K \) slightly decreases to 110 K upon 4% tensile strain while it
is strongly enhanced up to 200 K upon the compressive strain. The
strain-induced \( f-c \) hybridization change is also clearly observed in
the momentum-resolved spectral function as shown in Fig. 4c–e.
Not only the Kondo resonance state from \( j = 5/2 \) state at \( E_F \) but also	hose from \( j = 7/2 \) state at 0.3 eV are clearly enhanced upon the
compressive strain. The kink feature at 0.3 eV above \( E_F \) associated
with \( j = 7/2-c \) hybridization is very weak for 4% tensile strain but it
becomes much clear and stronger for compressive strain (dashed
circle in Fig. 4c). In general, the kink feature in the spectral function
is related to the formation of the Kondo resonance and the \( f-c \)
hybridization in heavy-fermion systems36.

Another notable aspect is band-dependent \( f-c \) hybridization.
This feature is most clearly observed in the case of \( -4\% \)
compressive strain. At the one-third point along the \( K-G \) line
(white arrow), two dispersive bands are hybridized with Ce 4f states.
However, the hybridization strengths of the two bands are quite
different. One band is strongly hybridized with the Ce 4f state
resulting in a large hybridization gap of 50 meV while the
hybridization gap from the other band is only 10 meV. This
difference arises from the different orbital character of the
conduction band. The left conduction band mainly comes from
Ce \( d_{z^2} \), whereas the right band mainly comes from Si \( p_x+p_y \) orbitals
(see Supplementary Fig. 2). The hybridization between Ce 4f and Si
\( p_x+p_y \) states is much stronger and sensitively affected by the strain
compared to the hybridization between Ce 4f and Ce \( d_{z^2} \) state.

Next, we investigate the surface doping effect on the Kondo
resonance state. There are two Ce-I layers above and below the
silicene layer within a monolayer as shown in Fig. 5a. By using the
virtual crystal approximation, we control the number of electrons
of the upper iodine layer, mimicking the surface doping situation.
The calculated Ce 4f orbitals occupancy of each upper and lower
Ce layer is presented in Fig. 5b. Regardless of surface doping, Ce 4f
electron occupancy remains constant for both Ce layers. However,
and lower Ce layer depending on surface doping. To answer this question, we construct a phase diagram as shown in Fig. 6, by calculating the inverse DOS (1/\(A_\omega(\omega = 0)\)) depending on strain. c–e Momentum-resolved spectral function for −4%, 0%, 4% strain at 10 K.

Fig. 4 Strain effect on Kondo resonance state. a Ce 4f spectral function, \(A_\omega\) at 10 K depending on the strain. b Temperature evolution of \(A_\omega(\omega = 0)\) depending on strain. c–e Momentum-resolved spectral function for −4%, 0%, 4% strain at 10 K.

Fig. 5 Surface doping effect on Kondo resonance state. a Schematic picture of surface doping. b Ce 4f occupancy of upper and lower Ce layer depending on surface doping. c \(A_\omega(\omega = 0)\) of the upper and lower Ce layer depending on surface doping. They are normalized by pure CeSi monolayer value.

Fig. 6 Phase diagram of Ce-based materials. Inverse density of state at \(E_F\) (x-axis) and inverse quasiparticle lifetime \(\Gamma\) (y-axis) of CeSiI in comparison with well-known heavy-fermion systems. Both quantities are normalized by the CeCoIn\(_5\) values.

Phase diagram and ground state prediction

An important remaining question is how sensitive the ground state of the CeSiI monolayer can be controlled by strain or surface doping. To answer this question, we construct a phase diagram as shown in Fig. 6, by calculating the inverse DOS (1/\(A_\omega(\omega = 0)\)) at \(E_F\) (x-axis) and inverse quasiparticle lifetime \(\Gamma = -\text{Im}\Sigma(0)\) (where \(Z^{-1} = m'/m = 1 - \text{Re}\Sigma(\omega)/\text{Im}\Sigma(\omega)\) (y-axis) based on PM DFT +DMFT calculations (at \(T = 46\) K) and in comparison with well-known Ce-based heavy-fermion materials as shown in Fig. 6. Both quantities of each material are normalized by those of CeCoIn\(_5\) for better comparison. The materials having PM heavy-fermion ground states are denoted by circles while those having magnetically ordered ground states are denoted by diamonds, respectively. Note that CeCoIn\(_5\) is located in the close vicinity of the QCP and magnetically ordered CeRhIn\(_5\) undergoes a superconducting transition only above 2 GPa, indicating the proximity to the QCP. Interestingly, the well-known Ce-based materials are well classified with respect to the CeCoIn\(_5\) point. The typical PM heavy-fermion systems are located below the CeCoIn\(_5\) point and magnetically ordered systems are located above the CeCoIn\(_5\). The PM ground state of CeRhIn\(_5\) under 7 GPa is also well captured.

Along with the well-known Ce-based compounds, CeSiI monolayers with and without external perturbation (strain, surface doping, and Si substitution) are also displayed with black and open squares in Fig. 6. For the surface doping case, only the \(\Gamma\) and 1/\(A_\omega(\omega = 0)\) of upper layer Ce are shown in the figure because those of lower layer...
Ce do not change upon surface doping on the upper iodine layer. CeSiI monolayer is located above the CeColn point, indicating a magnetically ordered ground state and it is consistent with the recent experimental result on bulk CeSiI[^11]. It is rather located closer to CeIn3 and CeCu2Ge2. Although CeRhIn5, CeIn3, and CeCu2Ge2 have magnetically ordered ground states, they have been intensively considered as AFM heavy-fermion systems since their ground state can be easily turned into PM heavy-fermion states by external pressure or chemical doping[^14]-[^17]. Therefore, CeSiI should also be located not far from the QCP, and its ground state can be easily tuned by external perturbations like CeIn3 and CeCu2Ge2.

As we have discussed above, compressive strain and surface hole doping strongly enhance the Kondo resonance peak of the CeSiI monolayer. With 4% compressive strain, it is located right below the CeColn, indicating a possible quantum phase transition to PM heavy-fermion state. With 0.1 surface hole doping, it moves further below CeColn and is located near the well-known PM heavy-fermion materials CeColn and CeCu2Si2. Therefore, CeSiI monolayer is a promising 2D vdW heavy-fermion candidate and its ground state can be tuned by strain or surface doping (or I substitution by Te), providing alternative ways to explore the QCP, magnetism, and unconventional superconductivity.

**DISCUSSION**

The interesting points of this new type of heavy-fermion system are low dimensionality and frustration originating from its own triangular lattice. During the last couple of decades, the degree of quantum fluctuation has been studied in addition to the Kondo coupling and RRK interaction. This quantum fluctuation can be enhanced by both lattice frustration and geometrical low-dimensionality. Together with the Kondo coupling strength, this new microscopic quantity (i.e., frustration) enables the construction of a 2D parameter space, zero-temperature global phase diagram[^38]-[^39]. So far, a complete understanding of this global phase diagram is still lacking.

By enhancing spatial dimensionality to reduce the quantum fluctuation, the cubic Ce$_3$Pd$_2$Si$_6$ was studied[^40]. One can tune Ce$_3$Pd$_2$Si$_6$ from a small Fermi surface (FS) AFM state, through a large FS AFM phase, into a large FS PM state. Recently, it is reported that geometrically frustrated CePdAl hosts a stable quantum critical phase, possibly PM small FS phase. YbRh$_2$Si$_2$[^41] and $\beta$-YbAlB$_4$[^42] also show similar quantum phase transitions. Although they do not have geometrical frustration, their quasi-2D crystal structure may enhance the quantum fluctuation and lead to a similar quantum phase transition trajectory on the global phase diagram.

However, an understanding of the microscopic processes remains missing. CeSiI monolayer can provide a new platform to study the global phase diagram. Due to its low dimensionality and lattice frustration, one can study the upper part of the global phase diagram, where the quantum fluctuation is strong. Besides, this 2D vdW heavy-fermion system has additional control parameters. In addition to magnetic field and pressure (uniform strain), uniaxial strain can be an external tuning parameter in the CeSiI monolayer. By inducing anisotropy via uniaxial strain, frustration can be controlled, providing a unique opportunity to explore the entire global phase diagram.

Another interesting aspect is the critical behavior of resistivity near this QCP with low dimensionality or frustration. In the epitaxially grown CeIn3/LaIn3 superlattice, the quantum phase transition trajectory within the 2D limit was observed by reducing the superlattice period. Near the QCP, the resistivity shows $T$-linear behavior deviating from the Fermi liquid behavior $\rho(T) = \rho_0 + AT^\alpha$, where $\alpha = 2$. This linear $T$-dependence ($\alpha = 1$) is also in contrast to the resistivity behavior observed near the pressure-induced QCP in the bulk CeIn3 ($\alpha = 1.5$[^18]). YbRh$_2$Si$_2$ also shows $T$-linear resistivity near the QCP[^43], while $\beta$-YbAlB$_4$ recovers the Fermi liquid behavior ($\alpha = 2$) at a very low temperature[^44]. Therefore, it would be interesting to study the resistivity behavior in the CeSiI monolayer to further understand the strange metallic behavior in the global phase diagram.

Finally, the CeSiI monolayer is a new building block to study emergent phenomena in vdW heterostructures. Although 17T/1H-TaS$_2$ heterostructure and twisted trilayer graphene can also be 2D heavy-fermion building blocks, delicate preparation is required since those heavy-fermion states are not intrinsic. It would be interesting to study vdW heterostructures which consist of CeSiI monolayer and other 2D vdW components, including magnetic materials. Although the epitaxially grown Kondo superlattice, such as CeColn/CeIn3 and CeColn/CeRhIn5, have been used to study the dimensionality effect and the interactions between heavy-fermion state (or superconducting state) and magnetism (bosonic excitations[^22]-[^23]), CeSiI monolayer provides more degrees of freedom to make various interfaces in combination with other 2D vdW materials or substrates.

To summarize, we have investigated 2D vdW heavy-fermion systems from the experimentally known bulk compounds by using DFT+DMFT calculations. Among them, CeSiI is the most promising candidate. We have found that the Kondo coupling strength of CeSiI does not change upon exfoliation and can be easily controlled by strain and surface doping. Finally, we have predicted the ground state of the CeSiI monolayer in response to external perturbations. Although the CeSiI monolayer has a magnetically ordered ground state, we found that the compressive strain and surface hole doping strongly enhance the Kondo coupling strength, indicating a possible quantum phase transition to PM heavy-fermion state. Our result thus suggests that the CeSiI monolayer can be a genuine 2D vdW heavy-fermion system and provide a new playground to study the QCP and emergent phenomena in vdW heterostructures.

**METHODS**

**DFT and DFT+DMFT calculations**

We first performed DFT open-core calculation using the Vienna ab-initio Simulation Package (VASP) for the screening process[^44]. The experimental crystal structure of 32 lanthanide materials employed from the Inorganic Crystal Structure Database (ICSD) was used in the calculations. For structural relaxations of the strained CeSiI cases, DFT open-core calculations with VASP were also employed since DFT calculations with 4f orbitals significantly overestimate $f$-$c$ hybridization, resulting in a smaller lattice constant. DFT open-core calculation with vdW correction well reproduced the experimental crystal structure of CeSiI. The internal atomic positions were fully relaxed for biaxial strained case, while the CeSiI monolayer structure was just adopted for surface doping cases.

To study the $f$-$c$ hybridization properly, we employed a DFT+DMFT calculation as implemented in DFT+Embedded DMFT (eDMFT) Functional code[^45]. DFT calculations were performed by using WIEN2k[^46] and the correlation effect of Ce 4f orbitals is treated by a DMFT loop. The hybridization energy window from $-10$ to $10$ eV with respect to the $E_F$ was chosen and $U = 5$ eV and $J = 0.68$ eV were used for all Ce-based compounds. The continuous-time quantum Monte Carlo (CTQMC) solver was basically adopted for all calculations (Fig. 6). The temperature evolution of the Kondo resonance peak on the real axis was analyzed using vertex corrected one-crossing approximation (OCA) solver to avoid the ambiguity arising from analytic continuation, which is inevitable for the CTQMC solver (Figs. 3 and 4)[^45].

**DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request.
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AUTHOR CONTRIBUTIONS

B.G.J. conceived the work. B.G.J. performed the DFT and DFT+DMFT calculations and analyzed the calculation results together with C.L., J.H.S., and J.-X.Z. All authors discussed the results and participated in the writing of the manuscript.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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