# **Different metamagnetism between paramagnetic Ce and Yb isomorphs**

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To solve the puzzle of metamagnetic phenomena in heavy-fermion systems, we have compared paramagnetic isostructural Ce and Yb systems, CeNi<sub>2</sub>Ge<sub>2</sub> and YbNi<sub>2</sub>Ge<sub>2</sub>, both of which are located near a magnetic instability. The most intriguing result is the discovery of a metamagneticlike anomaly for isomorphic Ce and Yb paramagnetic systems from magnetization measurements in a pulsed magnetic field. Similar to other metamagnets, the metamagnetic transition fields for both compounds are well scaled by the temperature  $T_\chi^{\text{max}}$ , at which the magnetic susceptibility shows a maximum. In addition, for CeNi<sub>2</sub>Ge<sub>2</sub>, a peak of nonlinear susceptibility  $\chi_3$ appears at approximately  $T_\chi^{\text{max}}/2$ , as for other heavy-fermion metamagnets. In contrast, YbNi<sub>2</sub>Ge<sub>2</sub> shows only a sign change for  $\chi_3$  at  $T_\chi^{\text{max}}$ , as observed in itinerant metamagnets located near the ferromagnetic critical point. The metamagnetism of CeN<sub>12</sub>Ge<sub>2</sub> corresponds to a typical Kondo lattice system, whereas that of YbN<sub>12</sub>Ge<sub>2</sub> is similar to the nearly ferromagnetic itinerant systems. Other possibilities for the metamagnetic behavior of YbNi<sub>2</sub>Ge<sub>2</sub> are also discussed.

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#### **I. INTRODUCTION**

Metamagnetic phenomena in strongly correlated itinerant electron systems have attracted much attention for a long time. Since the discovery of a nonlinear increase in magnetization *M* of CeRu2Si2 at characteristic fields of *Hm* ∼ 7*.*8 T along the tetragonal *c* axis [\[1\]](#page-4-0), extensive experimental and theoretical studies have been performed. Metamagnetism of  $CeRu<sub>2</sub>Si<sub>2</sub>$ is regarded as a crossover rather than as a phase transition, and hence is referred to as a pseudometamagnetic transition. When the system is tuned to the antiferromagnetic (AFM) phase by expanding its volume by chemical pressure, the first-order metamagnetic transition from AFM to the polarized paramagnetic (PPM) phase takes place at  $H_c$  [\[2\]](#page-4-0). In crossing the critical pressure  $(p_c)$  at which the AFM transition temperature  $T_N$  is suppressed to zero, the metamagnetic transition for CeRu<sub>2</sub>Si<sub>2</sub> becomes crossover, e.g.,  $H_c \sim H_m$  [\[2,3\]](#page-4-0). Another example of metamagnetism is the field-induced paramagnetic (PM) to ferromagnetic (FM) transition for the itinerant electron systems located near the FM critical point [\[4,5\]](#page-4-0). When the system is in the PM phase beyond the FM critical endpoint, the metamagnetic transition changes to crossover, as observed in UCoAl [\[4,](#page-4-0)[6\]](#page-5-0). In many itinerant electron systems, such as PM heavy-fermion and nearly FM systems, metamagnetism only appears below  $T_{\chi}^{\text{max}}$ , where the magnetic susceptibility  $\chi$ is at a maximum. In addition,  $H_m$  is known to be proportional to  $T_{\chi}^{\text{max}}$  [\[4,](#page-4-0)[7\]](#page-5-0). These facts indicate that metamagnetism and the maximum in  $\chi$  are dominated by a single energy scale, i.e., they have the same origin.

To reveal more details of metamagnetism in heavy-fermion systems, we focused on certain aspects of the well-studied  $ThCr<sub>2</sub>Si<sub>2</sub>$ -type tetragonal Ce and Yb systems for the following aspects. In the tetragonal symmetry, the crystalline electric field (CEF) split the  $J = 5/2$  (7/2) of Ce<sup>3+</sup> (Yb<sup>3+</sup>) into three (four) Kramers doublets. When the CEF splitting energy  $\Delta_{\text{CEF}}$ , typically of the order of 100–200 K, is larger than the

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Kondo temperature  $T<sub>K</sub>$ , the degeneracy of the doublet ground state must be resolved by forming the magnetic order or heavyfermion state. The balance between  $T_K$  and  $\Delta_{\text{CEF}}$  can be tuned by composition or the external parameters, such as pressure and doping. From the literature [\[7\]](#page-5-0), only the hexagonal and cubic compounds of the PM Yb systems were known to show metamagnetism, for example, YbCuAl  $[8]$ , YbAgCu<sub>4</sub>  $[9]$ , YbCu<sub>5</sub> [\[10\]](#page-5-0), and YbT<sub>2</sub>Zn<sub>20</sub> (*T* = Co, Rh, and Ir) [\[11,12\]](#page-5-0). In this context, the discovery of new examples exhibiting metamagnetism among the PM Yb systems having tetragonal symmetry would be desirable. Given the above issues, we focus on the Ce and Yb isomorphs,  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and YbNi<sub>2</sub>Ge<sub>2</sub>. Both compounds crystalize in the tetragonal  $ThCr<sub>2</sub>Si<sub>2</sub>$ -type structure and have PM ground states like  $CeRu<sub>2</sub>Si<sub>2</sub>$ . It is quite rare that both Ce and Yb isomorphs have a PM ground state and therefore this comparison may shed light on the difference or similarity between the 4*f* electron and hole analogues.

 $CeNi<sub>2</sub>Ge<sub>2</sub>$  is believed to be located near the AFM instability. At low temperature, the electrical resistivity, specific heat, and magnetic susceptibility deviate from the Fermi-liquid description [\[13,14\]](#page-5-0). For example, the low-temperature specific heat divided by temperature exhibits a  $-\sqrt{T}$  dependence with large value of the coefficient of electronic specific heat,  $\gamma = 350 - 450$  mJ/mol K<sup>2</sup> [\[13–15\]](#page-5-0). The thermal Grüneisen parameter diverges as  $T \rightarrow 0$  [\[16\]](#page-5-0). Large  $\gamma$  value and the diverging of the Grüneisen parameter recall to mind  $Cer(u_2Si_2)$ [\[17\]](#page-5-0). Both CeNi<sub>2</sub>Ge<sub>2</sub> and CeRu<sub>2</sub>Si<sub>2</sub> have the large Grüneisen parameter exceeding 100 as encountered in many heavyfermion systems [\[18,19\]](#page-5-0). The temperature dependence of *χ* for the *H*||c axis features a broad maximum at  $T_{\chi}^{\text{max}} \sim 28 \text{ K}$ [\[20\]](#page-5-0) for CeNi<sub>2</sub>Ge<sub>2</sub>, whereas  $T_{\chi}^{\text{max}} \sim 10 \text{ K}$  for CeRu<sub>2</sub>Si<sub>2</sub> [\[1\]](#page-4-0). The Pd substitutions at the  $\tilde{N}$  sites of CeNi<sub>2</sub>Ge<sub>2</sub> induce AFM ordering, which indicates proximity to an AFM phase [\[21–23\]](#page-5-0). CeNi<sub>2</sub>Ge<sub>2</sub> was reported to exhibit metamagnetism at *H<sub>m</sub>*  $\sim$  42 T in free powdered samples [\[20\]](#page-5-0) and  $\sim$ 43 T in oriented powdered samples [\[24\]](#page-5-0). Because of the relatively high  $H_m$ , the details of metamagnetism for  $CeNi<sub>2</sub>Ge<sub>2</sub>$  are still unclear.

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FIG. 1. Temperature dependence of the magnetic susceptibility  $M/H$  of (a) CeNi<sub>2</sub>Ge<sub>2</sub> and (b) YbNi<sub>2</sub>Ge<sub>2</sub> in magnetic fields of 0.1 T applied along the *a* and *c* axes.

YbNi2Ge2 has the relatively large *γ* value of 136 mJ*/*mol K<sup>2</sup> [\[25\]](#page-5-0) and has an intermediate Yb valence of  $\sim$ 2.8 at low temperature [\[26\]](#page-5-0). Interestingly, *χ* shows a broad maximum at approximately 50 K, for both  $H \| a$  and  $c$  [\[25\]](#page-5-0). Magnetic ordering was observed above  $p_c = 5$  GPa [\[27\]](#page-5-0), at which the Yb valence remains noninteger [\[26\]](#page-5-0). Although the magnetic structure above  $p_c$  is still not known, an FM interaction is indicated from the magnetoresistance [\[27\]](#page-5-0). This fact infers that  $YbNi<sub>2</sub>Ge<sub>2</sub>$  is located near an FM critical point. This is strikingly different from CeNi<sub>2</sub>Ge<sub>2</sub>, which is located near an AFM instability. Moreover, their magnetic easy directions are different, as seen in the susceptibility curves (see Fig. 1). From scaling,  $H_m \sim T_{\chi}^{\text{max}}$ , the metamagnetic behavior for both  $\text{CeNi}_2\text{Ge}_2$  and  $\text{YbNi}_2\text{Ge}_2$  is expected to be captured using a pulsed magnetic field.

In this paper, we compare the metamagnetic behavior of paramagnetic  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and  $YbNi<sub>2</sub>Ge<sub>2</sub>$  obtained from magnetization measurements for fields up to 56 T using a pulse magnet. Both are located near their respective magnetic critical point: AFM for  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and FM for  $YbNi<sub>2</sub>Ge<sub>2</sub>$ ; the magnetic anisotropy is also different, i.e., the easy magnetization *c* axis for the former and the easy basal plane for the latter. The main observation here is that both compounds feature pseudometamagnetic magnetization anomalies when the field is applied along the easy magnetization axis or plane. In particular, YbNi<sub>2</sub>Ge<sub>2</sub> might be an example of a PM Yb compound with tetragonal symmetry exhibiting a metamagnetic behavior. Differences appear in their temperature evolutions of magnetization and nonlinear magnetic susceptibility.

### **II. EXPERIMENT**

Single crystals of  $CeNi<sub>2</sub>Ge<sub>2</sub>$  were prepared by the Czochralski method, and those of  $YbNi<sub>2</sub>Ge<sub>2</sub>$  were grown by the In-flux method [\[25\]](#page-5-0). The temperature dependence of magnetization at 0.1 T is measured using a commercial superconducting quantum interference device (SQUID) magnetometer. Pulsedmagnetic fields up to 56 T were applied using a nondestructive magnet with typical durations of ∼36 ms installed at the International MegaGauss Science Laboratory of the Institute for Solid State Physics at the University of Tokyo. Magnetization in pulsed fields was measured by the conventional induction method, using coaxial pick-up coils.

# **III. RESULTS AND DISCUSSIONS**

Figure 1 presents the temperature dependence of the magnetic susceptibility *M/H* for applied magnetic fields of 0.1 T along the tetragonal *a* and *c* axes for CeNi<sub>2</sub>Ge<sub>2</sub> and  $YbNi<sub>2</sub>Ge<sub>2</sub>$ . The overall trends are in good agreement with the previous reports [\[20,25,26\]](#page-5-0). From the Curie-Weiss fit above 100 K, the effective moment of  $CeNi<sub>2</sub>Ge<sub>2</sub>$ , estimated to be ∼3.0  $\mu$ <sub>B</sub> for both directions, is slightly larger than a previous result of 2.84  $\mu$ <sub>B</sub> [\[20\]](#page-5-0) and than the expected value of 2.54  $\mu$ <sub>B</sub> for free Ce<sup>3+</sup> ion. Although the CEF schemes are still controversial, the splitting energy between the excited and ground states was reported to be  $200-300$  K  $[28,29]$ , which is comparable to the Curie-Weiss fitting temperature range. To determine the effective moment precisely, the susceptibility measurements at higher-temperature regions above 300 K are needed. The Weiss temperature  $\Theta_{a(c)}$  for  $H \| a(c)$  is evaluated as  $-206$  K ( $-56$  K), which is in agreement with the literature [\[20\]](#page-5-0). For YbNi<sub>2</sub>Ge<sub>2</sub>, the effective moment is 4.5 (4.6)  $\mu_B$ , which is near 4.54  $\mu$ B of the Yb<sup>3+</sup> ion, and  $\Theta_{a(c)}$  is −79 K  $(-156 \text{ K}).$ 

CeNi<sub>2</sub>Ge<sub>2</sub> has a maximum in *M/H* at  $T_{\chi}^{\text{max}} \sim 30$  K for only *H* ||c, whereas for YbNi<sub>2</sub>Ge<sub>2</sub>, a maximum at  $T_\chi^{\text{max}} \sim 50 \text{ K}$ appears for both *H* directions. In addition, the susceptibility of  $CeNi<sub>2</sub>Ge<sub>2</sub>$  features an upturn at low temperatures. Assuming the anomalous peak corresponds to a density of state of quasiparticles at the Fermi energy, the increase in  $\chi$  can be reproduced phenomenologically [\[14\]](#page-5-0). The singularity in the density of states relates strongly to the non-Fermiliquid behavior, which can also reproduce the temperature dependence observed for the specific heat. In addition, the mode-mode coupling theory predicts critical exponents at the AFM quantum critical point giving a  $\chi \propto -T^{1/4}$  and a  $-T^{1/2}$  dependence for the specific heat [\[30\]](#page-5-0). Although the evaluation of the exponent from Fig. 1 is difficult because low-temperature data are absent, the upturn is a consequence of the proximity to the AFM critical point. The upturn is strongly suppressed with fields, as will be discussed later. In contrast, the susceptibility of  $YbNi<sub>2</sub>Ge<sub>2</sub>$  monotonically decreases below  $T_{\chi}^{\text{max}}$ , suggesting that YbNi<sub>2</sub>Ge<sub>2</sub> is far from the magnetic instability, in agreement with a previous report [\[27\]](#page-5-0).

Note also that by replacing the Ce site by Yb, the magnetic easy direction is switched from the *c* axis to the basal plane although the magnetic anisotropy  $\chi_a/\chi_c$  of less than 2 is quite small. Such changes in magnetic anisotropy were also seen for  $CeRh<sub>2</sub>Si<sub>2</sub>$  and  $YbRh<sub>2</sub>Si<sub>2</sub>$  [\[31,32\]](#page-5-0). The anisotropy change between  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and  $YbNi<sub>2</sub>Ge<sub>2</sub>$  may be due to their different CEFs [\[25\]](#page-5-0). With tetragonal symmetry, the magnetic anisotropy is mainly dominated by the  $B_2^0O_2^0$  term of the CEF Hamiltonian, and the easy *c* axis and the *ab* plane are realized for the negative and positive  $B_2^0$ , respectively [\[33\]](#page-5-0). The CEF parameter  $B_2^0$  is evaluated from the difference between  $\Theta_a$ and  $\Theta_c$ , specifically,  $B_2^0 = 10(\Theta_a - \Theta_c)/[3(2J - 1)(2J + 3)]$ [\[25,34\]](#page-5-0). Using the estimated  $\Theta_{a(c)}$  from the Curie-Weiss fits,  $B_2^0$  for CeNi<sub>2</sub>Ge<sub>2</sub> and YbNi<sub>2</sub>Ge<sub>2</sub> are, respectively, estimated as −23 and 5 K, consistent with their magnetic anisotropy,



FIG. 2. Magnetic field dependence of magnetization at 1.4 K of (a) CeNi<sub>2</sub>Ge<sub>2</sub> and (b) YbNi<sub>2</sub>Ge<sub>2</sub> for the  $H||a$  and *c* axes. The differential susceptibility *dM/dH* for each is also presented.

i.e., the easy *c* axis and *ab* plane for the former and the latter, respectively.

Our main finding in this work is the discovery of an example exhibiting metamagneticlike nonlinear magnetization curves in both isomorphic Ce and Yb compounds having a PM ground state. Figure 2 presents the magnetization curves  $M(H)$  at 1.4 K for CeNi<sub>2</sub>Ge<sub>2</sub> and YbNi<sub>2</sub>Ge<sub>2</sub> for applied fields along the  $a$  and  $c$  axes. CeNi<sub>2</sub>Ge<sub>2</sub> clearly exhibits metamagnetic behavior at  $H_m = 45$  T for the  $H||c$  axis, whereas for the  $H||a$  axis,  $M$  monotonically increases up to the highest fields. This anisotropic behavior is common among Ce PM metamagnets,  $Ceku_2Si_2$  [\[1\]](#page-4-0) and  $CeFe_2Ge_2$ [\[35\]](#page-5-0). Within experimental error, hysteresis is not observed over a field cycle. Moreover, note the *M*(*H*) behavior above  $H_m$ . In CeNi<sub>2</sub>Ge<sub>2</sub>, the linear extrapolation of the  $M(H)$  curve above  $H_m$  crosses the origin, which is also seen in powdered samples [\[20\]](#page-5-0). This is in strong contrast to the isostructural Ce metamagnets,  $Ceku_2Si_2$  [\[1\]](#page-4-0) and  $CeFe_2Ge_2$  [\[35\]](#page-5-0). The finite intercept in  $CerRu_2Si_2$  may reflect the strength of the spin polarization. When crossing  $H_m$ , CeNi<sub>2</sub>Ge<sub>2</sub> seems to change its PM character to a weakly polarized spin state.

The hole analog  $YbNi<sub>2</sub>Ge<sub>2</sub>$  exhibits magnetization upturn, inferring metamagnetic behavior in a tetragonal Yb paramagnet. Interestingly, the fields along both directions induce magnetization upturns, which may be consistent with the appearance of the peak in the susceptibility at almost identical  $T_{\chi}^{\text{max}}$ , and therefore the same energy scale governs the maximum of the susceptibility and metamagnetism. The anomaly is clear to see for the easy magnetization *a* axis at  $H_m \sim 40$  T than that for  $H \| c$ . Contrary to CeNi<sub>2</sub>Ge<sub>2</sub>, the nonlinearity of magnetization is very weak, as seen in the very broad peak of *dM/dH*, and *M* does not tend to saturate at least up to 56 T. The  $J = 7/2$  degeneracy is suggested by the susceptibility and the Kadowaki-Woods ratio considering the degeneracy [\[26,36\]](#page-5-0). Higher fields are necessary to saturate to the value  $4 \mu_{\rm B}$  for a free Yb<sup>3+</sup> ion.

Hereafter, we concentrate on the easy direction, specifically,  $H \| c$  and  $H \| a$  for CeNi<sub>2</sub>Ge<sub>2</sub> and YbNi<sub>2</sub>Ge<sub>2</sub>, respectively. Figure 3 presents the  $M(H)$  curves at various temperatures.  $H_m$ is insensitive to temperature, and with warming the *M* anomaly



FIG. 3. Magnetization curves at various temperatures for (a)  $CeNi<sub>2</sub>Ge<sub>2</sub>$  (*H*||c) and (b) YbNi<sub>2</sub>Ge<sub>2</sub> (*H*||a). For clarity, the data are offset by (a) 0.15 and (b) 0.2  $\mu_B$ /f.u. The dashed lines are extrapolations of the linear field dependence of magnetization,

suggesting the disappearance of metamagnetism near  $T_\chi^{\text{max}}$ .

becomes indistinct. The linearity of *M*(*H*) (highlighted by the linear guide lines near  $T_\chi^{\text{max}}$ ) indicates the disappearance of metamagnetism above  $T_{\chi}^{\text{max}}$ . In the inset of Fig. 4, the peak of  $\chi = dM/dH$  in CeNi<sub>2</sub>Ge<sub>2</sub> appears to disappear near  $T_{\chi}^{\text{max}}$ . The height of the differential susceptibility of  $CeNi<sub>2</sub>Ge<sub>2</sub>$  at  $H<sub>m</sub>$ is determined by  $\Delta \chi_m = \chi_m - \chi_0$ , where  $\chi_{m(0)}$  is the  $\chi$  at  $H =$ *H<sub>m</sub>* (0.1 T). In contrast to the strong temperature dependence of  $χ<sub>m</sub>$ ,  $χ<sub>0</sub>$  exhibits very little dependence.  $Δχ$  does not diverge at finite temperature, inferring a pseudometamagnetic transition. This behavior is also commonly observed in  $Ceku_2Si_2$  and  $CeFe<sub>2</sub>Ge<sub>2</sub> [1,35,37].$  $CeFe<sub>2</sub>Ge<sub>2</sub> [1,35,37].$  $CeFe<sub>2</sub>Ge<sub>2</sub> [1,35,37].$  $CeFe<sub>2</sub>Ge<sub>2</sub> [1,35,37].$ 

To extract more details, we replotted *M/H* as a function of temperature at various constant fields in Fig. [5,](#page-3-0) with the data from Fig. 3. We first take a look at the characteristics for  $CeNi<sub>2</sub>Ge<sub>2</sub>$ . With increasing field, the upturn at low



FIG. 4. Temperature dependence of the inverse peak height of the differential susceptibility of CeNi<sub>2</sub>Ge<sub>2</sub> at  $H_m$ ,  $\Delta \chi_m = \chi_m - \chi_0$ . The inset shows the temperature evolution of *dM/dH*.

<span id="page-3-0"></span>

FIG. 5. Temperature dependence of  $M/H$  of (a) CeNi<sub>2</sub>Ge<sub>2</sub> ( $H||c$ ) and (b) YbNi<sub>2</sub>Ge<sub>2</sub> ( $H||a$ ) at several constant fields. The broken lines in (a) are only a visual guide.

temperatures is strongly suppressed and becomes constant at least above 20 T. This indicates the recovery of the Fermi-liquid state.  $T_{\chi}^{\text{max}}$  shifts to a lower temperature and tends towards 0 K as  $H \to H_m$ . A similar field evolution of the temperature dependence of *M/H* was also reported for CeRu<sub>2</sub>Si<sub>2</sub> [\[38\]](#page-5-0).  $M/H$  increases further with increasing field and saturates above ∼50 T. Up to the highest studied fields  $H/H_m \sim 1.24$ , the suppression of  $M/H$  at low temperature seen in  $Ceku_2Si_2$  [\[38\]](#page-5-0) is not observed. Next, we take a look at  $YbNi<sub>2</sub>Ge<sub>2</sub>$ . The tendency is similar to  $CeNi<sub>2</sub>Ge<sub>2</sub>$ , i.e., the broad maximum shifts to lower temperature with increasing field. In contrast to  $\text{CeNi}_2\text{Ge}_2$ , however, the broad maximum of  $M/H$ in YbNi<sub>2</sub>Ge<sub>2</sub> does not disappear even above  $H_m = 40$  T.

For CeNi<sub>2</sub>Ge<sub>2</sub>,  $T_{\chi}^{\text{max}}$  is close in energy to the spectral linewidth of the AFM fluctuation obtained from inelastic neutron scattering [\[15,39,40\]](#page-5-0); unfortunately, a similar measurement for  $YbNi<sub>2</sub>Ge<sub>2</sub>$  is lacking. Whether the magnetic and/or valence fluctuation exist in  $YbNi<sub>2</sub>Ge<sub>2</sub>$  is important to know. For  $CeNi<sub>2</sub>Ge<sub>2</sub>$ , as for  $CeRu<sub>2</sub>Si<sub>2</sub>$ , the suppression of the AFM fluctuation at  $H_m$  drives  $T_\chi^{\text{max}}$  to zero [\[41\]](#page-5-0). Also, low-energy spin fluctuations of around 0.6 meV were found to play an important role in the non-Fermi-liquid behavior [\[40\]](#page-5-0). Therefore, this low-temperature behavior and the high-field pseudometamagnetic transition of  $CeNi<sub>2</sub>Ge<sub>2</sub>$  are decoupled, as discussed in Ref. [\[3\]](#page-4-0). A comparison of the thermal and magnetic Grüneisen parameters may resolve the above issues.

The notable differences between  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and  $YbNi<sub>2</sub>Ge<sub>2</sub>$ appear in the temperature dependence of the nonlinear susceptibility  $\chi_3$ . The field expansion of the magnetization is written as  $M(H) = \chi_1 H + \frac{1}{3!} \chi_3 H^3$ , where  $\chi_1$  is the uniform magnetic susceptibility and higher-order terms are neglected. Therefore, these values can be determined experimentally from the plot of  $M/H$  vs  $H^2$ : the intercept and slope correspond to  $\chi_1$  and  $\chi_3$ , respectively. CeRu<sub>2</sub>Si<sub>2</sub>, for example, shows a maximum in both quantities: temperature  $T_3^{\text{max}}$  corresponds to a peak in  $\chi_3$ that is below  $T_{\chi}^{\text{max}}$  [\[42\]](#page-5-0).

Figure 6 represents the temperature dependence of  $\chi_1$  and *χ*<sup>3</sup> for CeNi2Ge2 and YbNi2Ge2, respectively. The consistency between  $\chi_1$  obtained from the fit and  $M/H$  data measured



FIG. 6. Temperature dependence of  $\chi_1$  (left axis) and  $\chi_3$  (right axis) of (a) CeNi<sub>2</sub>Ge<sub>2</sub> for  $H||c$  and (b) YbNi<sub>2</sub>Ge<sub>2</sub> for  $H||a$ , respectively. Symbols and dotted lines are, respectively, fitted results and the  $M/H$  measured at  $\mu_0H = 0.1$  T.

at  $H = 0.1$  T is rather good for YbNi<sub>2</sub>Ge<sub>2</sub>. In contrast, the discrepancy is larger for  $CeNi<sub>2</sub>Ge<sub>2</sub>$  because of the strong field-dependent non-Fermi-liquid behavior in  $\chi(T)$  [\[14\]](#page-5-0). For CeNi<sub>2</sub>Ge<sub>2</sub>,  $\chi_3$  exhibits a maximum at  $T_3^{\text{max}} \sim 13$  K, whereas for  $YbNi<sub>2</sub>Ge<sub>2</sub>$ , it monotonically decreases with increasing temperature and becomes negative at around  $T_\chi^{\text{max}}$ .

Recently, Shivaram *et al.* pointed out that  $T_3^{\text{max}}$  is scaled by  $T_{\chi}^{\text{max}}$  in many heavy-fermion systems having a diverse type of metamagnetic transitions [\[43\]](#page-5-0). They proposed a simple two-level system model, i.e., an excited pseudospin of  $S_z = \pm 1$ separated from the  $S_z = 0$  ground state by a gap  $\Delta$  yielding the scaling  $T_3^{\text{max}} / T_{\chi}^{\text{max}} \sim 0.4$ . The peak structures of  $\chi_1$  and  $\chi_3$  are dominated by a single energy scale  $\Delta$ , which is also related to  $H_m$ .  $T_3^{\max}$  of CeNi<sub>2</sub>Ge<sub>2</sub> is near  $T_\chi^{\max}/2$ , following the scaling [\[43\]](#page-5-0).

In striking contrast,  $YbNi<sub>2</sub>Ge<sub>2</sub>$  does not show any peak structure in  $\chi_3$ . The positive  $\chi_3$  gradually decreases with increasing temperature and becomes negative at around  $T_\chi^{\text{max}}$ . The universality observed in many heavy-fermion compounds and CeN<sub>12</sub>Ge<sub>2</sub>, i.e.,  $T_3^{\text{max}} / T_{\chi}^{\text{max}} \sim 0.4$  [\[43\]](#page-5-0), is not valid for  $YbNi<sub>2</sub>Ge<sub>2</sub>$ . The same characteristic behavior, however, was reported in nearly FM itinerant electron metamagnets  $YCo<sub>2</sub>$  $[44]$  and TiBe<sub>2</sub> [\[45\]](#page-5-0), which were not taken into consideration in the literature [\[43\]](#page-5-0). The sign change of  $\chi_3$  at  $T_\chi^{\text{max}}$  was explained well using Landau theory including the spin fluctuations [\[5\]](#page-4-0). The Landau-type expansion uses *M* as an order parameter and is shown to describe trends for the (nearly) FM systems well. In contrast, for the AFM, the sublattice magnetization needs to be taken into account. Quite recently, the metamagnetism of  $CeRu<sub>2</sub>Si<sub>2</sub>$  and the related systems were phenomenologically explained using a Landau-type free energy for an AFM Ising system with two sublattices [\[46\]](#page-5-0). The good description for  $YbNi<sub>2</sub>Ge<sub>2</sub>$  using the usual Landau theory indicates that the metamagnetisms exhibited by  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and  $YbNi<sub>2</sub>Ge<sub>2</sub>$  may be different in origin. It was suggested that the FM interaction plays an important role in the magnetically ordered phase of  $YbNi<sub>2</sub>Ge<sub>2</sub>$  under pressure [\[27\]](#page-5-0). And thus,  $YbNi<sub>2</sub>Ge<sub>2</sub>$  is located near the FM critical point at ambient pressure, leading in the similarity to the nearly FM systems. Moreover, most of the pressure-induced magnetic phases of Yb-based systems with ThCr<sub>2</sub>Si<sub>2</sub> structure such as YbCu<sub>2</sub>Si<sub>2</sub> [\[47,48\]](#page-5-0), YbIr<sub>2</sub>Si<sub>2</sub> [\[49\]](#page-5-0),

<span id="page-4-0"></span>and  $YbRh<sub>2</sub>Si<sub>2</sub> [50,51]$  $YbRh<sub>2</sub>Si<sub>2</sub> [50,51]$  seem to be FM. If the ordered phase above  $p_c$  is FM, the field-induced first-order metamagnetic transition from PM to FM is expected near  $p_c$  and in the PM phase  $[4]$ . Moving from  $p_c$  to the PM side, the transition changes to crossover across the quantum critical endpoint, as found for UCoAl  $[6]$ . For YbNi<sub>2</sub>Ge<sub>2</sub>, the PM phase is stabilized with decreasing pressure, and thus the metamagnetic crossover may take place at ambient pressure. The broadness of the magnetization anomaly is because the FM critical point is located far away. Pressure experiments can verify this scenario; pressure moves  $YbNi<sub>2</sub>Ge<sub>2</sub>$  to  $p<sub>c</sub>$  and changes the metamagnetic anomaly from crossover to first-order transition. Of course, an experiment revealing magnetism above  $p_c$  is most desired. Because of a lack of other comparisons and experimental investigations of  $YbNi<sub>2</sub>Ge<sub>2</sub>$ , it is at present difficult to conclude whether metamagnetism has its origin in FM fluctuations.

We discuss other alternative scenarios of the metamagnetic behavior in  $YbNi<sub>2</sub>Ge<sub>2</sub>$ . The theory based on the Coqblin-Schrieffer model revealed a magnetization upturn for  $J = 7/2$ and reproduced the metamagnetic behavior in YbCuAl with a single energy scale; it also explained the maximum in the temperature dependence of magnetic susceptibility and specific heat  $[8,52]$ . In the calculation for  $T=0$ , the coefficient of  $H^2$  term of  $M/H$ ,  $\chi_3$ , was found to be positive, in agreement with our results [\[8\]](#page-5-0). Although there is no theoretical investigation of the temperature dependence of  $\chi_3$ ,  $\chi_3 = 0$  at  $T_\chi^{\text{max}}$ is at least expected with the disappearance of magnetization upturn. A further theoretical investigation of magnetization for  $J = 7/2$  at finite temperatures is desired. Metamagnetism in the valence crossover regime is theoretically known [\[53\]](#page-5-0), in which divergences were seen not only for valence but also magnetic susceptibility at the field-induced valence quantum critical point and thereby initiating FM fluctuations. Indeed, such metamagnetic behavior accompanied by a large valence change is confirmed experimentally in  $YbAgCu<sub>4</sub>$  [\[54\]](#page-5-0). This may give rise to a similarity between valence changed metamagnets and nearly FM itinerant metamagnets. The very broad *M*(*H*) anomaly indicates weak magnetic and valence fluctuations; YbNi<sub>2</sub>Ge<sub>2</sub> has a relatively high  $p_c \sim 5$  GPa [\[27\]](#page-5-0). The evaluation of the field dependences of valence and volume deserves further attention so as to understand the metamagnetic behavior observed in  $YbNi<sub>2</sub>Ge<sub>2</sub>$ .

Recently, field-induced Lifshitz transitions featuring magnetization anomalies have also been reported, for example, in CeRu<sub>2</sub>Si<sub>2</sub> [\[55\]](#page-5-0) and YbRh<sub>2</sub>Si<sub>2</sub> [\[56,57\]](#page-6-0). The magnetization anomaly of the latter is a kink rather than a step [\[58\]](#page-6-0). For both compounds, the effective mass is reduced across the transition. Notably, the Lifshitz transition is not necessarily accompanied by a magnetization anomaly and a suppression of effective mass, as observed in CeIrIn<sub>5</sub> [\[59\]](#page-6-0). For YbNi<sub>2</sub>Ge<sub>2</sub> though, excluding the Lifshitz transition as the origin of the magnetization upturn is not possible at present. In this regard, Fermi surface studies across  $H_m$  gain some importance and urgency.

Also it is unclear at present whether the metamagnetic behavior of  $YbNi<sub>2</sub>Ge<sub>2</sub>$  is a specific case or a more general case of PM Yb systems having a tetragonal lattice. Finding other examples of such systems exhibiting similar properties and having a susceptibility maximum and easy-plane anisotropy would decide this issue.  $YbCu<sub>2</sub>Si<sub>2</sub>$ , which is located near the FM phase separated by  $p_c \sim 8$  GPa [\[48\]](#page-5-0), has a susceptibility maximum at  $T_{\chi}^{\text{max}} \sim 40 \text{ K}$  for  $H \| a \text{ [60]}$  $H \| a \text{ [60]}$  $H \| a \text{ [60]}$ . At least up to 50 T, however, no clear metamagnetic behavior is observed in YbCu<sub>2</sub>Si<sub>2</sub> in any direction, although the anisotropy  $\chi_c/\chi_a \sim 3$ and thus CEF are different from that in  $YbNi<sub>2</sub>Ge<sub>2</sub>$  [\[60\]](#page-6-0). Strong differences between  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and  $YbNi<sub>2</sub>Ge<sub>2</sub>$  appear in the temperature dependence of  $M/H$  near  $H_m$  and in  $\chi_3$  near  $T_{\chi}^{\text{max}}$ . To specify the characteristic features in YbNi<sub>2</sub>Ge<sub>2</sub>, determining whether other Yb metamagnets such as  $YbAgCu<sub>4</sub>$ [\[9\]](#page-5-0) and  $YbT_2Zn_{20}$  [\[12\]](#page-5-0) display a maximum or sign change in  $\chi_3$  would be of interest from a substitutional perspective. Although the substitution effect of Ce for Yb is not yet clear, the CEF scheme affects the anisotropy and seems to determine magnetic and/or valence fluctuations. A theoretical investigation considering CEF is strongly desired.

## **IV. CONCLUSION**

From the magnetization measurements in pulsed fields, we have observed an example of metamagnetic behavior in PM isomorphs  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and YbNi<sub>2</sub>Ge<sub>2</sub>. The behavior in both is a crossover rather than a phase transition. In contrast to a rather sharp pseudometamagnetic transition in  $CeNi<sub>2</sub>Ge<sub>2</sub>$ , the nonlinearity is very weak for  $YbNi<sub>2</sub>Ge<sub>2</sub>$ . Similar to other PM systems, the pseudometamagnetic fields can be scaled by the temperature corresponding to the susceptibility maximum. The temperature dependence of the linear and nonlinear susceptibility shows strong contrasts between  $CeNi<sub>2</sub>Ge<sub>2</sub>$  and  $YbNi<sub>2</sub>Ge<sub>2</sub>$ . The differences seem to depend on whether the systems are located near an AFM or FM critical point. Other possibilities, such as valence fluctuation and Lifshitz transition, are at present not excluded as the origin of the metamagnetic behavior of  $YbNi<sub>2</sub>Ge<sub>2</sub>$ . These findings are sufficiently intriguing to stimulate further investigations of metamagnetism in these systems.

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