Unusual ^{27}Al NMR shift in the Weyl-fermion systems LaAlGe and PrAlGe D. Tay,^{1,*} T. Shang^{2,3}, P. Puphal,^{3,4} E. Pomjakushina,³ H.-R. Ott,^{1,5} and T. Shiroka^{1,5,†}¹Laboratorium für Festkörperphysik, ETH Zürich, CH-8093 Zurich, Switzerland²Key Laboratory of Polar Materials and Devices (MOE), School of Physics and Electronic Science, East China Normal University, Shanghai 200241, China³Laboratory for Multiscale Materials Experiments, Paul Scherrer Institut, Villigen CH-5232, Switzerland⁴Max-Planck Institute for Solid State Research, D-70569 Stuttgart, Germany⁵Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

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LaAlGe and PrAlGe are members of the RAlGe series of compounds (with R a light rare-earth element), which have been shown to host Weyl fermions. By exchanging the rare-earth cation, RAlGe offers a remarkable tunability of its electronic properties thereby allowing to identify the electronic response of Weyl semimetals under varying temperature- and magnetic-field conditions. Through a comparative ^{27}Al NMR study of LaAlGe and PrAlGe , we show that the rare-earth element can also be used to tune the NMR response of Weyl fermions in the host. In both cases, we observe unusual NMR shifts that deviate significantly from those in ordinary metals, such as Al. In LaAlGe , we report the observation of a small negative shift, strongly varying with temperature, which can be explained by a recent theory of NMR shifts invoking Weyl fermions. In PrAlGe , instead, we report a giant positive NMR shift of up to 20% at 6 K, most likely due to the transferred hyperfine field from the Pr^{3+} $4f$ -electron local moments. We discuss the implications of our findings on future studies of Weyl-fermion systems.

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Introduction. The RAlGe compounds (with R a light rare-earth element, such as La, Ce, or Pr) have been shown to adopt a noncentrosymmetric LaPtSi -type crystal structure [1], thus, emerging as good candidates for hosting Weyl nodes in their electronic excitation spectra [2]. Recent theoretical work [3] has established that also compounds with local moments on the rare-earth site (in this case Ce or Pr) still qualify as Weyl semimetals. This is remarkable since cations carrying magnetic moments generally lead to complications at low temperatures where magnetic ordering breaks the time-reversal symmetry. Results of subsequent angle-resolved photoemission spectroscopy (ARPES) experiments confirmed these theoretical predictions in both LaAlGe [4] and PrAlGe [5]. In addition, results of measurements of the anomalous Hall and Nernst effects [6] provided indirect evidence of the Weyl nodes in PrAlGe , consistent with the theoretically calculated Berry curvature [6]. Hence, to date the existence of Weyl nodes in the electronic excitation spectra of the RAlGe family is well established.

The low-temperature magnetic properties of these compounds are definitely more elusive, since they depend on the details of their chemical composition and indirectly on the synthesis process [7]. Recent magnetotransport measurements on PrAlGe revealed that, instead of the simple ferromagnetic ground-state predicted by theory [3], the magnetic phase diagram of PrAlGe is more complex than anticipated [6]. The present state of understanding of the interplay among local

magnetic moments, external magnetic fields, and Weyl nodes is, thus, incomplete.

In this Rapid Communication, we first compare the unusual temperature dependence of the negative NMR shift in the ^{27}Al NMR spectra of LaAlGe with that in an ordinary metal, such as aluminum. The data differ significantly from those reported previously in Refs. [8,9] for other Weyl semimetals. In the first case, a positive shift of the ^{75}As resonance with a considerable temperature dependence was noted in TaAs [8]. In the second case, the ^{125}Te resonance line in WTe_2 revealed a negligible temperature dependence of the shift, and no information on the sign was provided [9]. The recorded shifts of up to 18% we report here for the ^{27}Al resonance in PrAlGe are significantly larger in magnitude, again exhibiting a considerable temperature dependence. Both features are most likely due to the influence of the transferred hyperfine fields caused by the interplay between the local Pr^{3+} $4f^2$ electronic moments and the fermions in the vicinity of the Weyl nodes in the RAlGe series.

Methods. First, PrAlGe polycrystalline rods for the floating-zone growth were cast by induction melting of the three starting elements Pr, Al, and Ge with a purity of 99.99% or better in a high-frequency ac electric field. The melting was followed by a sudden switch off of the field, causing the melt to drop into a cooled copper hearth. The rods were then transferred into a SCIDRE HKZ high-pressure high-temperature optical floating-zone furnace. Further details can be found in Ref. [7]. An oriented crystal of approximately 50 mg was finally cut for the NMR experiments. The LaAlGe polycrystalline sample was prepared by the arc-melting method. In both cases, phase purity was checked via x-ray

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FIG. 1. (a) Line shapes, (b) line shifts, and (c) linewidths of ^{27}Al NMR in nonmagnetic LaAlGe powder, measured in a 5-T applied magnetic field. The vertical line in (a) indicates the position of the reference Al NMR signal, whereas the dashed line in (b) is a fit to Eq. (4) and a guide to the eyes in (c). As shown in panels (b) and (c), in the nonmagnetic La case, neither the line shifts nor the widths exceed a few hundredths of a percent. Note that the ^{27}Al line in Al metal is practically temperature independent and located at +0.16% [10].

diffraction using a D8 Advance Bruker diffractometer with $\text{Cu } K\alpha$ radiation.

The magnetic susceptibility data shown below were recorded by means of a Quantum Design magnetic property measurement system. The NMR spectra were collected in a temperature range between 5 and 300 K via the spin-echo method by using $\pi/2$ - π rf pulses of 5 and 10 μs , respectively, with typical delays between the pulses of 20 μs . The ^{27}Al NMR reference signal was obtained by measuring the NMR resonance in metallic Al at room temperature and then by scaling its frequency to that of 1.1-m $\text{Al}(\text{NO}_3)_3$ in D_2O .

Experimental results. First we consider the ^{27}Al NMR spectra of paramagnetic LaAlGe (with a closed $5p^6$ configuration, La^{3+} has no magnetic moment). Since ^{27}Al is a spin-5/2 nucleus, ordinarily we expect to observe five NMR lines, reflecting the quadrupole interaction of the nonspherical nucleus with its electronic surroundings. It turns out that the quadrupole splitting is quite small and the satellite signals rather weak such that they are not discernible in the spectra shown in Fig. 1(a). Hence, the lines generally appear as Gaussians [Fig. 1(a)] with a small shift [Fig. 1(b)] and a small linewidth broadening [Fig. 1(c)] even at low temperatures. Also note that the NMR shifts are always negative (i.e., to the

left of the aluminium reference frequency, here represented by a dashed vertical line). Since the quadrupolar splitting is small and LaAlGe is weakly diamagnetic, the NMR behavior of LaAlGe is expected to be independent of the external magnetic field.

The situation is significantly different in the PrAlGe case. Here, the Pr^{3+} ions are known to carry a significant magnetic moment reflecting their $4f^2$ configuration. Because of the crystal-electric-field split multiplet [11,12], its magnitude increases with decreasing temperature and is likely to cause the observed significant broadening of the NMR line as well as its remarkable shift in frequency upon decreasing temperature. Indeed, in LaAlGe, the NMR shift between 6 and 300 K at 5 T is only approximately 6 kHz (0.01%). In PrAlGe, at the same applied field of 5 T, we observe a huge NMR shift, up to almost 10 MHz (20%)! [see Fig. 2(b)]. This giant NMR shift is accompanied by a significant lineshape broadening of up to around 2 MHz [Fig. 2(c)] at the lowest temperature covered in this study and is related to the onset of correlations among Pr^{3+} moments.

For comparison, we show the same type of data obtained in an applied magnetic field of 7 T. As in 5 T, the same huge NMR shift of more than 10 MHz persists [see Fig. 2(b)],

FIG. 2. (a) Line shapes, (b) line shifts, and (c) linewidths of ^{27}Al NMR in a magnetic PrAlGe crystal aligned along the c axis, measured in an externally applied field of 5 and 7 T (the line shapes at 7 T, not shown, are similar to those at 5 T). The vertical line indicates the position of the reference Al NMR signal, whereas the dashed lines are fits or guides to the eye. Note the giant shift of the NMR lines in panel (b) and the significant line broadening below 100 K in panel (c).

FIG. 3. (a) Inverse susceptibility, (b) Curie-Weiss-type $K_s(T)$, and (c) Clogston-Jaccarino plots showing the NMR shifts in a PrAlGe single crystal (with the field applied along the c axis) vs temperature and magnetic susceptibility, respectively, as measured at 5 T. Note the coinciding datasets in panels (a) and (b) under different applied magnetic fields and the similar $\Theta_W \sim 32$ -K values (T -axis intercepts). The dashed lines in (a) and (b) represent fits to the Curie-Weiss law. The fit in (c) highlights the linear relationship between the magnetic susceptibility and NMR line shift.

likewise accompanied by a large broadening of the NMR linewidth [see Fig. 2(c)]. As exemplified in Figs. 3(a) and 3(b), both $\chi(T)$ and $K(T)$ are only weakly affected by the external magnetic field. The Clogston-Jaccarino plot $K(\chi)$, involving data obtained at 5 T as displayed in Fig. 3(c), clearly reveals that $K(T)$ is dominated by $\chi(T)$ also for this type of material. Finally we note that $K(T)$ at both 5 and 7 T is represented by a Curie-Weiss-type fit with the same Θ_W temperature of 31.7 K. This value is practically identical to $\Theta_W = 32$ K, calculated from fitting the magnetic susceptibility data $\chi(T)$ shown in Fig. 3(a).

Discussion. Of prime interest here is a comparison of the experimental data of the ^{27}Al NMR lineshift in LaAlGe with those measured in Al metal [13] where, incidentally, $K(T) > 0$. As mentioned in the caption of Fig. 1(b), on the chosen scale, $K(T)$ in Al metal is represented by a horizontal line above the baseline $K = 0\%$. This is indeed very different from the LaAlGe dataset displayed in the same figure.

In order to interpret our lineshift data we employ the theory outlined in Refs. [14,15] where the anomalous hyperfine coupling of nuclear spins in Weyl metals is discussed. The Hamiltonian describing the spin-dipole part of the hyperfine coupling is identical for common and Weyl metals. The essential difference is that between the orbital parts. In ordinary metals, the orbital component of the electronic susceptibility describes the interaction between the nuclear spins and the orbital motion of electrons [see Eq. (1)]. In the case of Weyl fermions, instead, the orbital term is replaced by the spin of the Weyl quasiparticles [here denoted by the Pauli matrices $\sigma = (\sigma_x, \sigma_y, \sigma_z)$]. According to Ref. [14], the respective interactions are given by

$$\mathcal{H}_{\text{HFI}}^{\text{orb}} = \frac{\mu_0}{4\pi} \hbar \gamma_n g \mu^* \mathbf{I} \frac{\mathbf{r} \times \mathbf{p}}{\hbar r^3} \quad \text{for free electrons,} \quad (1)$$

$$\mathcal{H}_{\text{HFI}}^{\text{orb}} = \frac{\mu_0}{4\pi} \hbar \gamma_n e v_F \mathbf{I} \frac{\mathbf{r} \times \boldsymbol{\sigma}}{r^3} \quad \text{for Weyl fermions.} \quad (2)$$

Here, v_F is the Fermi velocity, μ^* is the renormalized chemical potential, g is the electron g factor, and μ_0 , $\hbar$, k_B , and e are physical constants.

In ordinary metals, the orbital component $H_{\text{HFI}}^{\text{orb}}$ is usually assumed to be temperature independent and, thus, included in the temperature-independent NMR shift K_0 . In Weyl semimetals, instead, $H_{\text{HFI}}^{\text{orb}}$ has an implicit temperature dependence due to σ , which is related to the temperature-dependent susceptibility. Consequently, in Weyl semimetals, we can decompose the Hamiltonian into a temperature-independent and a temperature-dependent part: $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}'(T)$. After some calculations [15], $\mathcal{H}'(T)$ can be expressed as

$$\mathcal{H}'(T) = e v_F \boldsymbol{\sigma}(T) \cdot \mathbf{A} + \frac{g \mu_B}{2} \boldsymbol{\sigma}(T) \cdot \mathbf{B}, \quad (3)$$

where $\mathbf{A}$ denotes the magnetic vector potential and $\mathbf{B}$ denotes the magnetic field. Note that, besides the usual Zeeman energy term, proportional to $\boldsymbol{\sigma}(T) \cdot \mathbf{B}$, there is also an additional term, proportional to $\boldsymbol{\sigma}(T) \cdot \mathbf{A}$. After some algebraic manipulation, the temperature-dependent NMR shift due to the Weyl fermions can be expressed as [15]

$$K(\mu, T) \approx \frac{\mu_0 e}{4\pi^2 \hbar} \left[\frac{g \mu_B}{\hbar v_F} \mu - \frac{e v_F}{3} \ln \left(\frac{W}{\max[|\mu|, k_B T]} \right) \right]. \quad (4)$$

Here, $W \gg \mu$ is a sharp high-energy cutoff regularizing the theory, whereas μ denotes the chemical potential away from the Weyl point. Note that μ contains an implicit temperature dependence (see Ref. [15] for details). The most salient feature of Eq. (4) is that at high temperatures ($k_B T \gg \mu$), the Knight-shift K is dominated by the $-\ln(W/k_B T)$ term, which gives rise to a *negative shift* regardless of the μ value.

The NMR shifts in LaAlGe can be explained qualitatively by this theory. From Fig. 1(b), it is clear that $K_s(T)$ in LaAlGe, although small, is *qualitatively different* from the free-electron-type $K_s(T)$ in the Al metal [13]. Therefore, we propose that the observed NMR shifts in LaAlGe are indeed due to the Weyl node features.

Based on ARPES measurement results [4], it was concluded that LaAlGe exhibits three classes of Weyl nodes, denoted by W_1 , W_2 , and W_3 respectively. The W_2 nodes are

FIG. 4. (a) Energy-momentum dispersion along the k_y direction as calculated from ARPES data for LaAlGe and PrAlGe [4,5]. The arrows indicate the W_2 and W_3 Weyl nodes located in proximity of the Fermi level. (b) Knight shift vs temperature for typical values of the chemical potential $\mu_{T=0}$ as resulting from Eq. (4). For convenience, we neglect the constant term and choose typical coefficients for the term proportional to μ and the logarithmic term, i.e., $K(\mu, T) = 100\mu + \ln(\max[|\mu|, k_B T])$. The Knight shift vs temperature for LaAlGe is shown in Fig. 1(b). The fit to Eq. (4) gives $\mu_{T=0} = 16.74$ meV with $\mu = 0$ corresponding to the chemical potential of the Weyl point.

type-II Weyl nodes, whereas the W_1 , W'_3 , and W''_3 nodes are of type I. In particular, the W_2 Weyl nodes are located almost exactly at the Fermi level [see Fig. 4(a)], whereas the W_1 , W'_3 , and W''_3 Weyl nodes are about 60, 110, and 130 meV above the Fermi level, respectively. Considering the very low energies involved in a typical NMR experiment (about tens of μeV), we may assume that the NMR response is due exclusively to the W_2 nodes.

NMR shifts due to a Weyl node in the typical ± 40 -meV range are illustrated in Fig. 4(b) where we plot Eq. (4) for selected values of $\mu_{T=0}$. We note that the shift at room temperatures is typically *negative*. As the temperature decreases, the NMR shift is increasingly dominated by μ and, hence, as expected from Eq. (4), it becomes less negative. Due to the implicit temperature dependence of μ , at intermediate T values, the Knight shift vs the temperature curves change with varying $\mu_{T=0}$. Nevertheless, the typical Knight shifts at a single Weyl node are predominantly negative, turning positive only at unphysically large $\mu_{T=0}$ values. In LaAlGe, in particular, a fit with $\mu_{T=0} = +16.74$ meV provides a fairly good agreement between the experiment and the theory [see Fig. 1(b)]. Given the relatively high chemical potential of +16.74 meV, the Fermi surface is rather far from the Weyl point. The satisfactory fit by means of Eq. (4) suggests that, under these circumstances, the system is less sensitive to the details of the dispersion relation near the Weyl point. At the current level of experimental precision, we do not observe any additional effects arising from the type-II classification of the Weyl node in LaAlGe.

On the other hand, in PrAlGe, the giant NMR shift cannot be accounted for by Eq. (4) only. Although it is certain that Weyl nodes exist also in PrAlGe—theoretical models [3] and ARPES results [5] agree that there are at least two (W_3 and W_4) Weyl nodes within +20 meV from the Fermi surface [Fig. 4(a)]—the giant NMR shift we observe in PrAlGe is *orders of magnitude* larger than the $\sim 0.1\%$ shifts observed in most Weyl systems studied so far (see, e.g., Refs. [8,9,15]). Clearly, the character of those shifts is qualitatively different from that observed in PrAlGe. The dominant contribution to Eq. (4) is the negative logarithmic term, which is *opposite* in

sign to the NMR shift observed in PrAlGe. The fact that the shifts at 5 and 7 T follow the same temperature dependence above the Curie temperature [see Fig. 3(b)], rules out the splitting of energy bands due to Landau diamagnetism [8].

As shown in Fig. 3(a), PrAlGe exhibits the typical $\chi(T)$ features of rare-earth compounds with 4*f*-electron local moments on regular lattice sites. On the other hand, our $\chi(T)$ data for LaAlGe indicate a weak diamagnetic response across the entire covered temperature range. Therefore, the ^{27}Al NMR resonance shift $K(T)$ in PrAlGe is expected to be dominated by the transferred hyperfine field [16,17] due to the Pr^{3+} ions. At elevated temperatures and at different applied magnetic fields, this contribution overshadows a possible influence of the Weyl-type quasiparticles on $\chi(T)$.

As mentioned above, the close connection between $K(T)$ [see Fig. 3(b)] and $\chi(T)$ is confirmed by the Clogston-Jaccarino-type plot shown in Fig. 3(c) where data recorded in a magnetic field of 5 T are shown as an example. The magnitude of the NMR shifts that we obtain in PrAlGe is consistent with the typical magnitudes of NMR shifts in compounds known to possess transferred hyperfine fields. Typical values of the transferred hyperfine field in Pr compounds are in the range of 1.3 to 13.6 T [16,17], thus, giving rise to correspondingly large NMR shifts. For instance, PrP shows a maximum NMR shift of 12% [17], whereas PrAl₂ exhibits an even higher NMR shift of 17.5% [18]. This implies that the very large shifts observed in our case are not a typical feature of Weyl-type metals but rather a consequence of the local-moment transferred hyperfine interactions with the itinerant quasiparticles. As such, they appear to be similar in either conventional metals or in Weyl-type semimetals.

In conclusion, we presented a comparative ^{27}Al NMR study of the Weyl semimetal systems LaAlGe and PrAlGe. The NMR shifts in LaAlGe can be described by an anomalous hyperfine coupling near the Weyl point. On the other hand, the magnetic PrAlGe exhibits a giant NMR shift of up to 20% at 5 T, which is absent in nonmagnetic LaAlGe. Thus, in order to identify the typical features of Weyl-type semimetals, the presence of local moments on the regular lattice sites of a chosen material should be avoided.

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