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<tr>
<td>Citation</td>
<td>Physical Review B, 95(19), p.195121_1-195121_8</td>
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<td>Text Version</td>
<td>Publisher's Version</td>
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<tr>
<td>DOI</td>
<td><a href="https://doi.org/10.1103/PhysRevB.95.195121">https://doi.org/10.1103/PhysRevB.95.195121</a></td>
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T dependence of nuclear spin-echo decay at low temperatures in YbRh$_2$Si$_2$

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(Received 31 December 2016; revised manuscript received 13 April 2017; published 10 May 2017)

The authors report $^{29}$Si nuclear spin-echo oscillations and decay measurements on a single crystal of YbRh$_2$Si$_2$. These quantities are found to be $T$ independent from 300 K down to $\sim$20 K, showing, however, a strong $T$ dependence below 20 K. These results indicate that electronic states near the Fermi level are modified at low temperatures. The observed spin-echo oscillations can be interpreted with the Ruderman-Kittel and pseudodipolar interactions between nearest-neighbor Si nuclei driven by conduction electron scattering at the Fermi surface. Possible modifications to the Fermi surface at low temperatures are discussed.

DOI: 10.1103/PhysRevB.95.195121

I. INTRODUCTION

The field-induced quantum critical phase transition (QCPT) in YbRh$_2$Si$_2$ at a critical field $H_c$ is not the usual case of spin-density wave instability observed in Ce-based compounds [1,2], but instead is a candidate for the locally critical QCPT case [3–5]. A strong-coupling model has also been proposed to be relevant [6]. A sudden change of Fermi-surface volume has been suggested theoretically [5] in the vicinity of the locally critical QCPT. In fact such a change of Fermi surface has also been suggested by transport measurements [7–10]. In addition, the Fermi surface is easily modified under magnetic field far above $H_c$ through a Lifshitz-type transition [11–14]. Furthermore, the electronic properties are quite sensitive to the valence of Yb [15,16]. Recent angle-resolved photoemission spectroscopy (ARPES) measurements on YbRh$_2$Si$_2$ indicated no drastic change of Fermi surface near the QCPT with decreasing temperature down to 1 K [17]. In contrast, heavy quasiparticle formation with change of Fermi surface was observed below 50 K in ARPES measurements using a laser photon source [18]. As described above, the precise nature of the Fermi surface of YbRh$_2$Si$_2$ remains controversial.

In our previous papers [19,20], coexisting static Fermi-liquid (FL) and non-Fermi-liquid (NFL) states near the QCPT in YbRh$_2$Si$_2$ were brought to light by means of $^{29}$Si nuclear spin-lattice relaxation time ($T_1$) studies with $H \parallel a$ and $c$ axes. The latter results may be related to localized character and valence instability of 4$f$ electrons in YbRh$_2$Si$_2$.

In the present investigation, $^{29}$Si nuclear spin-echo decay and oscillations are measured in YbRh$_2$Si$_2$ in order to probe changes in the electronic structure as a function of temperature. Observed strong modification of these quantities, especially of oscillations below 20 K, indicates a modification of electronic states at the Fermi level.

Ruderman and Kittel showed that conduction electrons induce an indirect scalar interaction between neighboring nuclear spins in metals. This is now well known as the Ruderman-Kittel (RK) interaction [21]. Subsequently, Bloembergen and Rowland pointed out the existence of a similar pseudodipolar (PD) indirect nuclear spin-spin coupling [22]. At the present time, RK and PD interactions are the only well-established origins for ($I = 1/2$) nuclear spin-echo oscillations in metallic systems. These interactions are known to be sensitive to modifications of electronic states near the Fermi level, since they arise through hyperfine couplings with conduction electrons. In fact, previously observed spin-echo oscillation results have been interpreted based on the RK-PD model, and the character of the Fermi surface has been discussed [23–25]. More recently, a combination of NMR data and direct calculations has been employed to study indirect nuclear spin-spin couplings and the bulk electron states that give rise to them in the topological insulator Bi$_2$Se$_3$ [26].

The present results are interpreted in the framework of the RK-PD model, which is considered to be basically correct for heavy fermion systems, whereas the ingredients of indirect nuclear spin-spin coupling may be difficult to identify due to heavy conduction-band formation and the related complex nature of the hyperfine interactions. Tentatively, a possible modification of Fermi surface for the observed $T$ dependence of the spin-echo oscillation frequency is presented in this paper, based on a simplified picture.

By way of introduction, we note that the YbRh$_2$Si$_2$ lattice presents nearly ideal circumstances for measuring the total spin-spin coupling between nearest-neighbor (NN) $^{29}$Si pairs using NMR techniques. The crystal structure of YbRh$_2$Si$_2$ is shown in Fig. 1. The Si lattice sites in this structure occur in NN pairs—two pairs per unit cell separated by a vector $\vec{R}$ oriented along the $c$ axis—that are rather closer together than second, third, etc., neighbors. As a result, the NN spin-spin couplings play a major role in the $^{29}$Si ($I = 1/2$, $\gamma/(2\pi) = 845.77$ Hz/Oe) spin-echo decay process. There are other circumstances that were also key to the success of this investigation. First, the measured $^{29}$Si linewidth $\gamma \Delta H/2\pi \sim 9$ kHz reveals a modest level of inhomogeneous line broadening that is evidently a consequence of disorder. As will be clear in the technical discussion below, this broadening renders the spin-spin couplings among the $^{29}$Si completely static, a condition required for the observation of oscillatory modulation of spin echoes [24].

Second, isotopic enrichment of the $^{29}$Si nuclear species not only greatly increases the NMR signal amplitude, but
also gives rise to a substantial fraction of 29Si NN pairs that are in this case the sole source of spin-echo oscillations. At a concentration c, the fraction of NN pairs that are both occupied with 29Si is c², the fraction that are unoccupied is (1−c)², and the fraction that is singly occupied is 2c(1−c). Thus, the fraction of 29Si which have a NN spin, allowing them to contribute to the spin-echo oscillation amplitude, is simply c, which is 0.52 in the present case. The fraction without a NN spin is therefore 1−c. Actually, the spin-echo oscillation amplitude is ∼10 times smaller in sample with natural abundance of the 29Si, i.e., c = 0.047. By measuring the echo oscillation frequency it is possible to monitor the magnitude of their indirect spin-spin couplings and how they vary with temperature. Such measurements reveal significant changes in the electronic state at low temperatures that occur in the vicinity of the QCPT in this system.

II. NMR SPIN-ECHO DECAY

The spin-echo amplitude m(2τ) decays with increasing time interval τ between the first (π/2) and second (π) excitation pulses as a result of nuclear spin-spin interactions (Fig. 2).

\[
\frac{m(2\tau)}{m(0)} \approx \exp\left\{ -\frac{2\tau}{T_{1L}} - \frac{1}{2} \left( \frac{2\tau}{T_{2G}} \right)^2 \right\} \\
\times \left[ f_{\text{non}} + f_{\text{osc}} W(2\tau) \right],
\]

(1)

where \( f_{\text{osc}} \approx c = 0.52 \) and \( f_{\text{non}} = 1 - f_{\text{osc}} \approx 0.48 \) are the estimated fractions of the echo signal that do and do not oscillate, respectively, during the echo decay. Also, \( 2\tau / T_{1L} \) is an exponential relaxation term in the exponent, represented here by spin-lattice relaxation time \( T_1 \) data from previous studies [19,20,27]. \( T_{2G} \) is a Gaussian model spin-spin relaxation time, which approximately represents the collective relaxation effect of 29Si beyond the first neighbor shell. \( W(2\tau) \) is a fractionally oscillatory echo-modulation factor which is the main focus of our study. See Sec. V for more details.

As shown in Fig. 1, there is a single NN site for each Si. (In a previous paper [28] a case with four NN sites was erroneously discussed for this system.) Consistent with the RK-PD model (see Sec. VA) for the single NN site case, \( W(2\tau) \) is well fitted in the present case with a single oscillation frequency \( G_{a,c} \).

\[
W(2\tau) = \cos(2\pi G_{a}\tau) \quad \text{for } H \parallel a: \theta = \frac{\pi}{2},
\]

\[
W(2\tau) = \cos(2\pi G_{c}\tau) \quad \text{for } H \parallel c: \theta = 0,
\]

(2)

where \( \theta \) is the angle between the c axis and magnetic field.

III. EXPERIMENT

Single crystals of YbRh2Si2 were grown with the 29Si isotope enriched to \( c = 52\% \), improving the NMR sensitivity by a factor ∼11 by the In-flux method previously described [19]. High sample purity was confirmed by the small residual resistivity \( \rho_0 \sim 0.99 \mu\Omega\text{cm} \) and the large RRR value \( \rho(300\,\text{K})/\rho(2\,\text{K}) = 104 \). No magnetic impurity phase was detected in magnetic susceptibility measurements. The antiferromagnetic phase transition was observed near 80 mK in zero field.

A single-crystal specimen \((6 \times 3 \times 0.1\,\text{mm}^3)\) was mounted in a 4He cryostat with an NMR pickup coil. The present measurement has been done under an applied field \( H = 7.2\,\text{T} \). In YbRh2Si2, \( T_1 \) [19,20] and \( T_{2G} \) decrease with decreasing applied field. Thus, the applied field \( H = 7.2\,\text{T} \) employed for the present study was chosen so that \( T_1 \) and \( T_{2G} \) would be long enough to allow precise determination of echo oscillation frequencies. Using a standard \( \pi/2 - \pi \) pulse sequence, the spin-echo intensity \( m(2\tau) \) so generated was measured as a function of the time \( \tau \) between the pulses to record the spin-echo decay (see Figs. 2 and 3). Here, a typical \( \pi/2 \) pulse width was 4–5 μsec. Since the resonance linewidth is rather narrow (e.g., ∼9 kHz at 7.2 T), all nuclear spins in the spectrum were quite uniformly excited by the radio frequency pulses used. The pulse repetition time \( t_{\text{rep}} \) was adjusted to be much longer than the previously determined \( T_1 \) [19,20].

Spin-echo oscillations due to RK and PD interactions occur if local inhomogeneous broadening is larger than the indirect RK and PD interactions [24]. This is clearly the case, since the observed inhomogeneous linewidth ∼9 kHz at 7.2 T is much larger than the estimated \( G_{a,c} \). Moreover, the echo...
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FIG. 3. $2\tau$ dependence of $m(2\tau)/m(0)$ for $H \parallel a$ and $c$ axes at 7 and 100 K. Solid lines are obtained using LS curve fitting for Eqs. (1) and (2). Previously measured $T_1$ values for $H \parallel a$ and $c$ axes are adopted respectively here [19,20]. Parameters obtained from the fits, i.e., $1/T_2^{\text{Ga,c}}$ and $G_{\text{Ga,c}}$, are presented in Fig. 5.

relaxation process appears to be caused entirely by reversal of neighboring nuclear moments by the rephasing pulse. That is clear, because when the rephasing pulse angle is small (see Fig. 4), $T_2^{\text{Ga}}$ becomes very long. Thus, any genuinely dynamic transverse relaxation is very long, indeed.

IV. SPIN-ECHO DECAY RESULTS

A. $T$ dependence of spin-echo decay

Figure 3 shows the spin-echo decay results for YbRh$_2$Si$_2$ with $H \parallel a$ and $c$ axes. As shown in the figures, the experimental results can be fitted well using Eqs. (1) and (2), taking account of fractional occupation for $^{29}$Si. The $T$ dependence of $G_a$ and $G_c$, as well as that of $T_{2Ga}$ and $T_{2Gc}$, have been measured for $H \parallel a$ and $H \parallel c$, respectively. For this analysis, previously determined $T_1$ values for $H \parallel a$ and $c$ axes have been adopted from [19] and [20], respectively.

Figure 4 shows spin-echo decay results with different refocusing-pulse angles for $H = 7.2$ T $\parallel c$ axis at 100 K. As the refocusing pulse width decreases, the spin-echo decay becomes slower, indicating that the spin-echo decay process is dominated by pulse modulation of static couplings, except for the (weak) $T_1$ relaxation process. In this situation, the echo is relaxed by the reorientation of neighbor spins by the refocusing (second) pulse [29]. These measurements confirm that the echo oscillations are not affected by other dynamical fluctuations, and $T_{1L}$ can be replaced approximately by $T_1$. In addition, if the spin-echo intensity is optimized with a $\pi/2 - \pi$ pulse sequence, the echo-decay curve is independent of pulse width, i.e., strength of radio-frequency field, on condition that all nuclei are uniformly excited.

Figures 5(a) and 5(b) show the $T$ dependence of $1/T_2^{\text{Ga,c}}$. From $T = 300$ K down to $\sim 20$ K, $G_a$ and $G_c$ are almost $T$ independent. In contrast, a strong $T$ dependence appears below 20 K, indicating that the indirect spin-spin coupling is $T$ dependent at low temperatures. As the indirect nuclear spin-spin coupling is induced through (heavy) conduction bands, the present observation is naturally explained by a modification of electronic states at the Fermi level below 20 K.

Actually, a shoulder structure has been found below $\sim 20$ K in the $T$ dependence of the Hall coefficient [30]. In addition, for the Seebeck coefficient $S$, $d^2S/dT^2$ was reported to become negative below $\sim 20$ K [31], also $T$ dependence of $1/\tau(\omega)$ starts to deviate from the usual Drude behavior below 20 K in optical conductivity measurements [32]. The inset to Fig. 5(a) shows the $T$ dependence of $1/T_1T$ below 100 K [19,20]. It seems that $d^2(1/T_1T)/dT^2 \sim d^2\text{Im} \chi/dT^2$ also becomes negative below $\sim 20$ K ($\text{Im} \chi$ is the dissipative term of the dynamic susceptibility). These behaviors can be related to the observed modifications of $G_{a,c}$, as these coefficients are sensitive to the electronic state at the Fermi level. Further
B. T dependence of $1/T_{2G}$

As shown in Fig. 5(a), $1/T_{2G}$ increases with decreasing $T$, especially for the case of field $H \parallel a$. We suggest that temperature changes in $G_{a,c}$ should be accompanied by similar changes in the more distant coupling parameters [see $J_{0j}$ in Eq. (7)]. Because $T_{2G}$ decay is driven by inversion of neighboring $^{29}$Si spins by the refocusing pulse as in Fig. 4, we believe this process to be dominated by couplings with the 12 neighbor sites that lie within a range of $4 \AA \leq r_{ij} \leq 5 \AA$. These provide a distribution of couplings that are random in sign which, by the central limit theorem, yield an essentially Gaussian broadening effect. Such a distribution, when Fourier transformed, yields a result of the form of Eq. (1). With more detailed modeling of the indirect couplings, it should be possible to estimate $T_{2G}$ in this fashion and relate its temperature dependence to that of $G_{a,c}$. In the meantime the form of this result is known, and its qualitative behavior is seen to be similar to that of $G_a$ and $G_c$, as expected.

V. ANALYSIS BASED ON THE RK-PD MODEL

In this section, the observed $T$ dependence of $G_{a,c}$ is interpreted based on the RK-PD model. At first, $T$ dependence of RK interaction $J$ and PD interaction $B$ is estimated from $G_{a,c}$. Then the $T$ dependence of $J$ and $B$ is assumed to be attributed to a modification of the Fermi surface. Based on this simplified picture, possible modification of the Fermi surface is discussed.

A. Modulation of the spin-echo decay ascribed to RK and PD interactions

It is well known that the RK and PD interactions can induce oscillations in the spin-echo decay curve [24,25]. In metals the indirect RK interaction between nuclear spins $I_i, I_j$ occurs through second-order scattering of conduction electrons [21], taking the scalar form

$$H_J = \sum_{i>j} J_{ij} I_i \cdot I_j.$$  \hspace{1cm} (3)

In addition and in contrast to the RK term, the PD interaction is a tensor quantity [25],

$$H_B = \sum_{i>j} B_{ij} (I_i \cdot I_j - 3I_i^z I_j^z),$$  \hspace{1cm} (4)

$$B_{ij} = \frac{1}{2} b_{ij} (3\cos^2\theta_{ij} - 1),$$  \hspace{1cm} (5)

where $\theta_{ij}$ is the angle between $\vec{r}_{ij}$ and the applied magnetic field, $\vec{r}_{ij}$ being the radius vector from the $i$th to the $j$th nucleus. $b_{ij}$ is the effective coefficient for an indirect dipolar form of interaction $B_{ij}$ between the $i$th and $j$th nuclear spins. Thus, the PD interaction depends on the direction of the applied field.

A complete formulation of the decay and modulation of free-induction and spin-echo signals has been given by Alloul and Froidevaux (AF) [25]. As will be demonstrated below, the $T_2$ process for this system is driven exclusively by nuclear-spin inversion of neighbor spins having significant spin-spin coupling, as described by (3) and (5). In such a case and with spins $I = 1/2$, the AF calculation gives the following exact result for the echo decay wave form of a nuclear spin “0” at $T = 0$.
the origin,

\[
m(2\tau) = \sum_{\lambda} \Pi_{j>0} \cos(2\pi \tilde{J}_{0j} \tau),
\]

where index zero refers to the relaxing $^{29}\text{Si}$ spin at the origin, and \(\sum_{\lambda}\) indicates a normalized sum over all configurations of neighbor spins given a concentration \(c\) of $^{29}\text{Si}$, and where

\[
h \tilde{J}_{0j} = J_{0j} + (b_{0j} - h^2 \gamma^2 \gamma_{0j}^3)(1 - 3\cos^2 b_{0j}).
\]

The second term in the parentheses is the classical dipole-dipole interaction between spins at the 0th and \(j\)th sites, which is of comparable magnitude to the PD coupling.

The first cosine factor in the indicated product in Eq. (6) represents the effect of $^{29}\text{Si}$ NN pairs in the YbRh$_2$Si$_2$ lattice, to which we ascribe all of the oscillatory behavior observed in our spin-echo decay data. We carry out the statistical sum over NN sites and introduce special parameter definitions for NN sites as follows: Thus, \(hJ \equiv J_{01}, hB \equiv b_{01}, 2\pi B_{c1} \equiv h\gamma^2 R^{-3}, R \equiv r_{01}, \) and \(\theta \equiv \theta_{01}\), whereupon Eq. (6) can be rewritten as

\[
m(2\tau) = (2c^2 \cos[2\pi(J + (B - B_{c1}) - 3\cos^2 \theta)]\tau)
+ 2c(1 - c) \sum_{\lambda} \Pi_{j>0} \cos(2\pi \tilde{J}_{0j} \tau).
\]

In the latter expression, the prefactor cosine term \(\propto 2c^2\) describes oscillations from NN pairs that are expected to modulate the echo decay wave form, while the \(2c(1 - c)\) term is the portion of the wave form from singly occupied pair sites that does not oscillate. The summation \(\sum_{\lambda}\) describes the part of \(m(2\tau)\) that is represented in Eq. (1) by the factor \(\exp[-0.5(2\tau/T_{2G})^2]\). Below, the \(2c^2\) term in the prefactor of Eq. (8) is fitted to spin-echo oscillation data to extract experimental values of \(J\) and \(B\) as a function of temperature. Meanwhile, the oscillatory modulation factor in Eq. (1) (square brackets) is identified as the quantity in angle brackets in Eq. (8) that includes the singly occupied NN pair sites that make no contribution to echo oscillations.

In this frame, the obtained \(G_{a,c}\) [see Eq. (2)] of YbRh$_2$Si$_2$ may be expressed,

\[
G_a = \pm (J + B - B_{c1}); \theta = \frac{\pi}{2},
\]

\[
G_c = \pm (J - 2B + 2B_{c1}); \theta = 0.
\]

While technically there appear to be four possibilities in Eq. (9), physically only two are allowed, namely both (+) and both (−). Which of these is applicable can only be decided by other considerations, but is not required for a fit to our experimental data. The classical dipolar parameter \(B_{c1}\) is estimated to be 347 Hz using \(R = 2.39\) Å in YbRh$_2$Si$_2$.

**B. T dependence of \(J\) and \(B\)**

As shown in Fig. 5(c), \(J\) and \(B\) are estimated using relations \(J = \pm(2G_a + G_j)/3\) and \(B = \pm(G_a - G_j)/3 + B_{c1}\) where the double signs are coordinated. There are two cases, case I where \(J = (2G_a + G_j)/3\) and \(B = (G_a - G_j)/3 + B_{c1}\), and case II, where \(J = -(2G_a + G_j)/3\) and \(B = (G_a - G_j)/3 + B_{c1}\). It should be noted that this behavior can be field dependent [12], though details of this effect are beyond the scope of this paper. For both cases I and II, \(|J|\) increases with decreasing \(T\). The amount of change of \(J\) and \(B\) below 20 K, i.e., \(\Delta J \equiv |J(2 K) - J(20 K)|\) and \(\Delta B \equiv |B(2 K) - B(20 K)|\), show a similar dependence on \(T\), suggesting that the origin of these changes may be similar. In contrast, \(B\) increases for case I, but for case II it decreases with decreasing \(T\). This difference is discussed below. In addition, \(1/T_{2G}\) increases with decreasing \(T\), indicating that changes of \(J\) and \(B\) lead to changes in the couplings \(\tilde{J}_{0j}\) for more distant neighbors as well.

As \(J\) and \(B\) are sensitive to the character of the Fermi surface [22], the present results can be attributed to a modification of the Fermi surface below 20 K. In addition to the field-induced Lifshitz transition previously confirmed in YbRh$_2$Si$_2$ [11–13], one possible origin for this modification may be a temperature-induced Lifshitz transition, such as that observed, e.g., in WTe$_2$ [33].

On the other hand, modification of the Fermi surface due to a heavy quasiparticle band formation has been indicated below 50 K in previous ARPES measurements [18]. Thus, this heavy quasiparticle band formation is another possible origin for the modification of \(J\) and \(B\). It is also consistent with the previously estimated Kondo temperature 25 K [31].

In contrast with the foregoing, no overall modification of the Fermi surface has been observed in recent ARPES measurements [17]. If the latter outcome holds up, then attribution of the observed \(T\) dependence of \(J\) and \(B\) to modifications of the Fermi surface would appear to contradict such a result. However, considering the following point, we think that there is not necessarily a contradiction here. The RK interaction between local Si sites is considered to occur via particular regions of the Fermi surface. Thus, the present results do not necessarily indicate an overall modification of the Fermi surface. The characteristics of such regions are discussed below.

**C. Change of \(J\) and \(B\) due to Fermi-surface modification**

The RK coupling \(J\) is related to electronic states at the Fermi level as follows [21,22]:

\[
hJ = C_0(\Delta_0^2)_{E_F} F_0(k_F).
\]

\[
\Delta_0^2 = u_k(r_i)u_k^*(r_i)u_k(r_j)u_k^*(r_j) = u_k(r_i)^2u_k(r_j)^2,
\]

\[
F_0(k_F) = 2k_F \cos(2k_F R) - \sin(2k_F R),
\]

where \(r_i\) and \(r_j\) are NN Si sites; \(e^{ikr}u_k(r)\) is a typical Bloch function indexed by \(k\); \(k_F\) is the Fermi wave vector; \(C_0\) is a parameter that is considered to be nearly \(\)T independent (see the Appendix); \(k\) and \(k'\) are occupied and unoccupied states, respectively, near \(k_F\). As \(r_i\) and \(r_j\) are identical sites in the present case, Eq. (11) can be simplified. \(\Delta_0^2\) may correlate with the density of states at the Fermi level. \(F_0(k_F)\) represents the oscillatory character of the RK interaction as shown in Fig. 6.

On the other hand, for the parameter \(B\), expansion of the Bloch states in spherical harmonics [34] may be useful. Thus,

\[
u_k(r) = u_k^0(r)P_0 + ikc_1u_k^1(r)P_1(\cos\theta_{kr})
- k^2c_2u_k^2(r)P_2(\cos\theta_{kr}) + \cdots,
\]

\[
P_l(\cos\theta_{kr}) \equiv \sum_{m=-l}^{m} (-1)^m P_l^m(\cos\theta_{kr}),
\]
where the $c_{1,2} > 0$ are constants determined near $k = 0$. $P_m$ is the associated Legendre function; $\theta_{k_r}$ is the angle between $\mathbf{k}$ and $\mathbf{r}$; $u_1^k(r)$ is the coefficient for the $P_1$ term ($P_0 = 1$). The $P_0$, $P_1$, and $P_2$ terms represent the $s$, $p$, and $d$ orbital character of $u_k(r)$, respectively. Then $B$ is expressed in terms of $u_1^k(r)$ and $u_2^k(r)$ as

$$
hB = C_1(\Delta_{1}^2)_{E_s} F_1(k_F) + C_2(\Delta_{d1}^2)_{E_s} F_2(k_F) + C_3(\Delta_{d2}^2)_{E_s} F_3(k_F), \quad (15)
$$

$$
F_1(k_F) \equiv \int_{0}^{k_F R} (\sin x - x \cos x)(\cos x - x \sin x) dx, \quad (16)
$$

$$
F_2(k_F) \equiv \int_{0}^{k_F R} \cos x(-x^2 + 3 \sin x - 3 x \cos x) dx, \quad (17)
$$

$$
F_3(k_F) \equiv \int_{0}^{k_F R} \sin x(-x^2 + 3 \cos x + x \sin x) dx, \quad (18)
$$

$$
\Delta_{1}^2 \equiv u_1^k(r_1)u_1^k(r_1) \int_{0}^{r_1} u_1^k(r)u_1^k(r) r^{-1} dr, \quad (19)
$$

$$
\Delta_{d1}^2 \equiv u_1^k(r_1)u_2^k(r_1) \int_{0}^{r_1} u_1^k(r)u_2^k(r) r^{-1} dr, \quad (20)
$$

$$
\Delta_{d2}^2 \equiv u_1^k(r_1)u_2^k(r_1) \int_{0}^{r_1} u_2^k(r)u_2^k(r) r^{-1} dr, \quad (21)
$$

where $r_1$ is the radius of the Wigner-Seitz sphere; $C_{1-3}$ are constants which are considered nearly $T$ independent (see the Appendix). It should be noted that estimates of $C_{1-3}$ are beyond the scope of this paper.

Figure 6 shows the $k_F$ dependence of $F_{1,2,3}(k_F)$. Regardless of $k_F$, $F_3$ is positive, whereas $F_1$ and $F_2$ are negative. As regards the $T$ dependence of $B$ for cases I and II, the positive third $F_3$ term in Eq. (15) becomes dominant, compared with the negative first ($F_1$) and/or second ($F_2$) terms in Eq. (15) below $20 \text{ K}$, or vice versa.

Although certain Fermi surfaces have not yet been observed in de Haas-van Alphen (dHvA) effect measurements at $T = 30 \text{ mK}$ and field values above 16 T [16], the observed $k_F$ is $\approx 1 \sim 3 \times 10^{10} \text{ m}^{-1}$, leading to $Rk_F = 2-7$. Even if this value of $Rk_F$ is relevant for the present result, it is difficult to tell the sign of $J$ from $F_0$ in Fig. 6 considering the uncertainty of the value of $Rk_F$. Nevertheless, if the overall Fermi surface is unchanged [17], the value of $k_F$, and thus the values of $F_{0-3}$ are considered to be essentially unchanged in the paramagnetic state down to $70 \text{ mK}$. Therefore, the present $T$ dependence for $B$ can be attributed to the fact that the dominance among the $\Delta_{d2}^2$, $\Delta_{d1}^2$, and $\Delta_{d1}^2$ is $T$ dependent below $20 \text{ K}$ at $7.2 \text{ T}$ for certain bands which give efficient contributions to the electron density of states at the Si site. As $\Delta J$ and $\Delta B$ show similar $T$ dependence, the $T$ dependence of $\Delta_{d1}^2$ for $|J|$ may be due to the modifications of $\Delta_{d2}^2$ and $(\Delta_{p}^2, \Delta_{d1}^2)$, which is natural since $\Delta_{d1}^2$ correlates with the total density of states at the Fermi level.

At the Si site, $\Delta_{p}^2$ may be mainly due to the atomic $2p$ orbital of Si; in contrast, hybridization between the $2s, 2p$ orbitals of Si, $4d$ orbitals of Rh, and $4f$ orbitals of Yb is necessary to induce a $\Delta_{d1}^2$ term.

The present results can be attributed to that the Fermi surface that depends on these hybridizations has a $T$ dependence.
below 20 K. If heavy quasiparticle band formation [18] is the case, $k_F$ can be $T$ dependent, which can induce a modification of $J$ and $B$ in addition to the mechanism described above. At present, unfortunately, more concrete changes of the Fermi surface cannot be specified from this result.

Since the present measurements reflect the local electronic state at the Si site, the observed change of electronic state could be largely local. This may be only weakly sensed by $k$-space probes, since a local change might only correspond to a small modification over a broad region of $k$ space. The absence of overall change of Fermi surface in the ARPES measurements [17] could be explained by this picture. In fact, the Fermi surface seems to appear differently, depending on the experimental probe up to now. This sort of complexity could be partly due to such a particular nearly localized state, which is considered to be related with the possibly exotic QCPT. As the observed spin-echo decay curve can be explained by the RK-PD model, the description via the RK-PD model appears to be valid for a heavy fermion system.

**ACKNOWLEDGMENTS**

We are grateful for stimulating discussions with H. Harima, D. Aoki, and K. Izawa. This work was supported by JSPS KAKENHI Grants No. 15H05884 (J-Physics) and No. 15K05152 and the REIMEI Research Program of JAEA.

**APPENDIX**

The constants in Eqs. (10) and (15) are expressed as follows [22]:

$$C_0 \equiv \frac{m'}{2\pi^3 \hbar^2 R^3} \left( \frac{16 \pi g_N \beta_N \beta}{3} \right)^2,$$

$$C_1 \equiv \frac{8m' c_1 c_1'}{15 \pi^3 \hbar^2 R^6} \left( \frac{16 \pi g(g_N \beta_N \beta)^2}{3} \right),$$

$$C_2 \equiv \frac{c_2}{4c_1 c_1'},$$

$$C_3 \equiv \frac{c_1}{c_2},$$

where $m'$ is the effective cyclotron mass of the electron; $g_N$ and $g$ are the $g$ values of nuclei and electrons, respectively; $\beta$ and $\beta_N$ are the electron and nuclear Bohr magnetons, respectively; the $c_{1,2}$ are constants in Eq. (13) for the $n_{\text{Fermi}}$ state. If we take $m'$ to be the electron rest mass, $C_0$ is found to be $2 \times 10^{-30}$ J ~ $3 \times 10^3$ Hz. Then, for tentative reasonable values $|F_0| = 1$ and $\langle \Delta_0^2 \rangle_{E_F} = 0.1$, $|J| = C_0 |F_0| \langle \Delta_0^2 \rangle_{E_F}$ is found to be $3 \times 10^3$ Hz, which is comparable with the observed $|J|$. On the other hand, $C_{1-3}$ cannot be estimated, since $c_{1,2}$ and $c_{1,2}'$ are unknown.

**VI. CONCLUSION**

The $^{29}$Si nuclear spin-echo oscillations and $T_{2G}$ show clear strong $T$ dependence at low temperatures in YbRh$_2$Si$_2$, indicating that the electronic state at the Fermi level starts to change below 20 K. This energy scale is consistent with other previous measurements, which may be characterized by the Kondo temperature 25 K. However, the corresponding change of Fermi surface may be peculiar in YbRh$_2$Si$_2$ due to a nearly localized electronic state, which is considered to be related with the possibly exotic QCPT. As the observed

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[21] M. A. Ruderman and C. Kittel, Phys. Rev. 96, 99 (1954); it should be noted that the RK interaction between nuclear moments here is different from the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between electron magnetic moments.