Ruderman-Kittel interaction between Si in URu$_2$Si$_2$

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Abstract. $^{29}$Si nuclear magnetic resonance (NMR) has been studied in a $^{29}$Si-enriched single crystal sample of URu$_2$Si$_2$. The spin-echo decay for applied field $H \parallel [110]$ and [001] directions has been measured at 50 K. A clear spin-echo decay oscillation is observed for both cases, reflecting the Ruderman-Kittel interaction between Si nuclei. Since the observed oscillation frequency depends on the direction of applied magnetic field, the Ruderman-Kittel and anisotropic pseudo-dipolar interactions may not be negligible in this compound. The origin of spin-echo decay oscillations is discussed.

1. Introduction
The heavy fermion compound URu$_2$Si$_2$ undergoes a phase transition [$1, 2, 3$] at $T_0 \sim 17.5$ K. Since the order parameter of the transition has not ever been clearly identified, such order has been termed “hidden order” [4] and has been studied intensively. In addition, the superconducting transition appears only in the hidden ordered state, indicating that the superconducting pairing glue can be related to fluctuations that occur within the hidden order. Previously, the hidden order and superconducting states have been studied by means of NMR Knight shift and spin-lattice relaxation measurements [5, 6, 7, 8]. Recently, we recognize that NMR spin-echo decay measurements are quite useful for investigating the electronic state of strongly correlated electron systems. It is well known that the spin-echo decay process oscillates due to the Ruderman-Kittel (RK) interaction [9, 10] between nuclei mediated by the conduction electrons. Thus, the modification of conduction bands can be measured via the spin-echo decay. Indeed, a strong change of spin-echo oscillation frequency is observed at low temperatures in YbRh$_2$Si$_2$ [11].

In the present study, we report data for the $^{29}$Si nuclear spin-echo decay in URu$_2$Si$_2$. The observed spin-echo decay curves show a clear oscillation, which we attribute to the RK interaction. The anisotropic oscillation frequency indicates that the contributions of the pseudo-dipolar and dipolar (PDD) interactions are not negligible.

2. Experimental
Experimental data have been presented and discussed in a previous report [6]. Single crystals were grown using the Czochralski method in a tetra-arc furnace under an argon gas atmosphere,

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as previously described. A single crystal sample with a nearly perfect cylindrical shape (1 mm
θ⊥[001]×3 mm∥[001]), with narrow facets on the (110) planes, has been used.

In the present study, high sample purity was confirmed by a residual resistivity of ~ 5 μΩcm
(RRR ~ 70). The resistivity was measured in a piece cut (2 × 0.5 × 0.5 mm³) from the same
single crystal used for NMR measurements.

The natural abundance of the NMR isotope ²⁹Si (I = 1/2, gyromagnetic ratio
29γ/2π = 0.84577 kHz/Oe) is only 4.7%. This has prevented highly accurate Si NMR
measurements in URu₂Si₂ in the past. For the present study, a single crystal sample with a
53% enriched ²⁹Si isotope has been prepared, improving the NMR sensitivity by a factor ~
11. As there are no quadrupolar interactions for I = 1/2 nuclei, ²⁹Si NMR spectra reflect only
magnetic shift and broadening effects.

A single crystal specimen was mounted in a ⁴He cryostat with an NMR pickup coil. Using a
standard π/2 − π pulse sequence, the spin-echo intensity m(2τ) so generated was measured as a
function of the time τ between excitation pulses (see Fig. 1). Here, a typical π/2 pulse width is
4 ~ 5 μsec. Since the resonance linewidth is quite narrow (e.g. ~ 1 kHz at 2.5 T along the [110]
axis in the paramagnetic state), all nuclear spins in the spectrum were quite uniformly excited
by the radio-frequency pulses used. The pulse repetition time t_rep was taken to be much longer
than the previously determined T₁ [12].

Figure 1. Schematic description of NMR spin-echo formation. A π/2 pulse rotates the nuclear
magnetic moment by 90°, a π pulse by 180°.

3. NMR Spin-echo decay
The spin-echo intensity m decays with increasing time period τ between the first (π/2) and
second (π) pluses due to several interactions between nuclei spins. Generally, the τ dependence
of m may be expressed;

\[
\frac{m(2\tau)}{m(0)} = \exp\left\{-\frac{2\tau}{T_{1L}} - \frac{1}{2}\left(\frac{2\tau}{T_{2G}}\right)^2\right\}\Omega(2\tau),
\]

(1)

where T₁L is the Lorentzian type relaxation. For the present case T₁L can be replaced by
the spin-lattice relaxation time T₁ determined previously [12]; T₂G is the Gaussian spin-spin
relaxation time. Ω(2τ) is the modulation factor which is under investigation.

3.1. Spin-echo decay modulation due to RK and PDD interactions
It is well known that the RK and PDD interactions can induce oscillations in the spin-echo
decay curve [13, 14, 15]. In metals the indirect RK interaction between nuclear spins Iᵢ, Iⱼ
occurs through second-order scattering of conduction electrons [9], taking the scalar form
\[ J_{ij} \mathbf{I}_i \cdot \mathbf{I}_j. \]  
(2)

In contrast to the scalar RK interaction (Eq. 2), the PDD interaction is a tensor quantity \[10],

\[ B_{ij}(\mathbf{I}_i \cdot \mathbf{I}_j - 3I_i^2 I_j^2), \]  
(3)

\[ B_{ij} = \frac{1}{2} b_{ij}(3\cos^2 \delta_{ij} - 1), \]  
(4)

where \( \vec{r}_{ij} \) is the radius vector between the \( i \)th and the \( j \)th nuclei, \( \delta_{ij} \) is the angle between \( \vec{r}_{ij} \) and the applied magnetic field, and \( b_{ij} \) is the effective coefficient for a dipolar-type interaction \( B_{ij} \) between the \( i \)th and \( j \)th nuclei. Thus, the PDD interaction depends on the direction of applied field.

A complete formula for the spin-echo decay modulation factor \( \Omega(2\tau) \) due to RK and PDD interactions is given by Alloul and Froidevaux \[15\]. As they have used polycrystalline samples, angle-integrated equations are given. Here, using their formulas, equations for the \( I = 1/2 \) case for a single crystal are presented. Considering the nearest neighbor (nn) sites among ions considered (i.e. Si for the present case),

\[ \Omega(2\tau) = \sum_{r=0}^{N} P_r \sum_{k=1}^{n_r} \rho_{k,r} \sum_{\alpha=1}^{r} \cos^2(2\pi \tau G_{\alpha,k}), \]  
(5)

\[ G_{\alpha,k} = J + B - 3B\cos^2\Theta_{\alpha,k}, \]  
(6)

\[ \sum_{k=1}^{n_r} \rho_{k,r} = 1, \]  
(7)

where \( J \) and \( B \) are the RK and PDD interactions between nn nuclear spins, respectively; \( N \) is the total number of nn sites; \( r \) corresponds to the number of occupied nn sites (i.e. with \(^{29}\)Si nuclear spins); \( P_r = c^r (1 - c)^{N-r} \) is the binomial coefficient; \( c = 0.53 \) is the concentration of \(^{29}\)Si isotope; \( n_r \) is number of possible configurations of occupied nn sites for a given \( r \), the configurations being indexed by \( k \), \( \rho_{k,r} \) is a distribution of situation \( k \) of \( r \) case; \( \Theta_{\alpha,k} \) is the angle between the applied magnetic field \( H \) and the direction to the \( \alpha \)th nn for the situation \( k \). It should be noted that contributions of unlike spins are negligible in the present case.

3.2. \( URu_2Si_2 \) case

For the case of \(^{29}\)Si in \( URu_2Si_2 \), there is only one nn site, namely along the \( c \) axis, i.e \( N = 1 \) (Fig. 2), as is also the case for \( YbRh_2Si_2 \). The RK and PDD interactions (i.e. \( J \) and \( B \)) are proportional to \( R^{-3} \) (\( R \) is distance between nn sites). Since the distance to four second nn sites (3.75\AA) is comparable with that of the nn site (2.35\AA) in \( URu_2Si_2 \), both cases are included here. The definition of angles \( \phi, \theta \) and \( \psi \) is given in Fig. 2.

3a) \( \Omega_{1st}(2\tau) \) for the first nn case

The nn case is simple since there is only one nn site (\( N = 1, n_r = 1 \)), which was already discussed previously for \( YbRh_2Si_2 \) \[11\] (note: definition of \( \theta \) in Ref. \[11\] corresponds to present \( \phi \)). There is a unique oscillation frequency \( G \) for \( \Omega_{1st}(2\tau) \). Thus,

\[ \Omega_{1st}(2\tau) = (1 - c) + c \cos(2\pi \tau G), \]  
(8)

\[ G \equiv \pm(J_1 + B_1 - 3B_1\cos^2\phi), \]  
(9)
where $J_1$ and $B_1$ are the RK and PDD interactions between the first nn sites, respectively. The double signs are coordinated. The signs in Eq. 9 cannot be determined by the present method; fortunately, they are not required in a fit to the data. For the nn site, $\Omega(2\tau)$ is independent of $\theta$ since the nn sites are located along the $c$ (\([001]\)) axis.

b) $\Omega_{2nd}(2\tau)$ for the second four nn case
The second four nn sites ($N = 4$) case is more complicated. All possible configurations $n_r$ for $r = 0 - 4$ cases should be considered. Based on Eq. 5, $\Omega_{2nd}(2\tau)$ may be expressed:

$$
\Omega_{2nd}(2\tau) = (1 - c)^4 + \\
2c(1 - c)^3\{\cos(2\pi\tau G_I) + \cos(2\pi\tau G_{II})\} + \\
c^2(1 - c)^2\{\cos^2(2\pi\tau G_I) + \cos^2(2\pi\tau G_{II})\} + \\
4c^2(1 - c)^3\{\cos(2\pi\tau G_I)\cos(2\pi\tau G_{II})\} + \\
2c^3(1 - c)\{\cos^2(2\pi\tau G_I)\cos(2\pi\tau G_{II}) + \cos(2\pi\tau G_I)\cos^2(2\pi\tau G_{II})\} + \\
c^4\{\cos^2(2\pi\tau G_I)\cos^2(2\pi\tau G_{II})\}.
$$

(10)

$G_{I,II}$ are defined as:

$$
G_I \equiv \pm\{J_2 + B_2 - 3B_2(\sin\psi\sin\phi\cos\theta + \cos\psi\cos\phi)^2\},
$$

$$
G_{II} \equiv \pm\{J_2 + B_2 - 3B_2(\sin\psi\sin\phi\sin\theta + \cos\psi\cos\phi)^2\}.
$$

(11)
Table 1. Values of $1/T_{2G}$ and $G$ obtained from fits based on Eqs. 1 and 8, along with previously determined $1/T_1$ values [12], which are adopted as $1/T_{1L}$.

|       | $1/T_{1L}$ (sec$^{-1}$) | $1/T_{2G}$ (sec$^{-1}$) | $G$(kHz) |
|-------|-------------------------|--------------------------|----------|
| $H$ || [001]      | 2.3                     | 452 ± 15 | 1.026 ± 0.02 |
| $H$ || [110]      | 17.1                    | 572 ± 11 | 0.583 ± 0.01 |

where $J_2$ and $B_2$ are the RK and PDD interactions with the second nn sites, respectively. The double signs are coordinated. The signs in Eq. 11 cannot be determined by the present method; fortunately, they are not required for a fit to the data. In URu$_2$Si$_2$, cos$\psi$ = 0.626. Compared to the first nn case, the second nn case contains many different oscillation frequencies. Note that Eq. 10 has 4-fold $\theta$ symmetry as expected.

Generally, the contribution from the nn site is dominant. If the second nn contributions are not negligible, $\Omega(2\tau)$ may be expressed [15];

$$\Omega(2\tau) = \Omega_{1st}(2\tau)\Omega_{2nd}(2\tau),$$

which is often a complicated function of $2\tau$.

4. Results and discussion

Figure 3. Spin-echo decay curves at 50 K for $H \parallel [001]$ ($\phi = 0$) and $H \parallel [110]$ ($\phi = 90^\circ$, $\theta = 0$) in URu$_2$Si$_2$. Solid lines are fitting curves based on Eqs. 1 and 8. Parameters obtained are presented in Table 1.

Figure 3 shows spin-echo decay data for $H = 2.5$ T || [001] and [110] directions at 50 K. The data obtained can be well fitted using Eqs. 1 and 8, indicating that the contributions from the second nn sites are almost negligible. It should be noted that no better fitting can be obtained using Eq. 12, indicating that the first nn contribution dominates the spin-echo decay at the Si site in URu$_2$Si$_2$. As the $G$ values obtained depend on the direction of field, certainly the PDD interaction should be considered in the present case. Based on Eq. 9 and $G$ values from Table 1, equations $\pm (J_1 - 2B_1) = 1026; \pm (J_1 + B_1) = 583$ are deduced. From these equations, $|J_1| = 731$
Hz and $|B_1| = 148$ Hz are obtained. If we consider the nuclear dipole-dipole contribution of 324 Hz in this enriched $\text{URu}_2\text{Si}_2$, the purely pseudo-dipolar interaction is estimated $148 + 324 = 472$ or $176$ Hz (see [11]).

5. Summary
In the present study, spin-echo oscillations have been observed in the paramagnetic state of $\text{URu}_2\text{Si}_2$. The results are well accounted for by the Ruderman-Kittel interaction. As the Ruderman-Kittel interaction is due to conduction electrons, the spin-echo oscillation frequency is considered to be sensitive to modifications of the Fermi surface in $\text{URu}_2\text{Si}_2$. This is a possible topic for future investigation.

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References
[1] Palstra T T M, Menovsky AA, van den Berg J, Dirkmaat A J, Kes P H, Nieuwenhuys G J, and Mydosh J A 1985 Phys. Rev. Lett. 55 2727.
[2] Schlabitz W, Baumann J, Pollit B, Rauchschwalbe U, Mayer H M, Ahlheim U and Bredl C D 1986 Z. Phys. B Condensed Matter 62 171.
[3] Maple M B, Chen J W, Dalichaouch Y, Kohara T, Rossel C, Torikachvili M S, McElfresh M W and Thompson J D 1986 Phys. Rev. Lett. 56 185.
[4] Tripathi V, Chandra P and Coleman P 2007 Nature Phys. 3 78.
[5] Kambe S, Tokunaga Y, Sakai H, Matsuda T D, Haga Y, Fisk Z and Walstedt R E 2013 Phys. Rev. Lett. 110 246406 and references therein.
[6] Kambe S, Tokunaga Y, Sakai H and Walstedt R E 2015 Phys. Rev. B 91 035111.
[7] Walstedt R E, Kambe S, Tokunaga Y and Sakai H 2016 Phys. Rev. B 93 045122.
[8] Hattori T, Sakai H, Tokunaga Y, Kambe S, Matsuda T D and Haga Y 2016 J. Phys. Soc. Jpn. 85 073711.
[9] Ruderman M A and Kittel C 1954 Phys. Rev. 96 99.
[10] Bloembergen N and Rowland T J 1955 Phys. Rev. 97 1679.
[11] Kambe S, Sakai H, Tokunaga Y, Hattori T, Lapertot G, Matsuda T D, Knebel G, Flouquet J and Walstedt R E 2017 Phys. Rev. B 95 195121.
[12] Kohori Y, Matsuoka K and Kohara T 1996 J. Phys. Soc. Jpn. 65 1083.
[13] Walstedt R E, Dowley M W, Hahn E L and Froidevaux C 1962 Phys. Rev. Lett. 8 406.
[14] Froidevaux C and Weger M 1964 Phys. Rev. Lett. 12 123.
[15] Alloul H and Froidevaux C 1967 Phys. Rev. 163 324.