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magnetic order (µ ∼ 0.02µB/U, T_N ∼ 17 K) and uncoventional superconductivity (T_c ∼ 1.2 K) [4].

Understanding the magnetic and superconducting behavior of this material has proven to be very challenging and has generated a substantial amount of work over the years (see e.g., Refs. [1–14] and references therein).

The transition at T_N is still puzzling. Although neutron diffraction (ND) experiments indicate antiferromagnetic order [15], the ordered-moment size is too small to account for macroscopic effects in the thermodynamic quantities [16]. Thus, many more studies have appeared with the goal of elucidating the nature of the true order parameter for this transition (see e.g., Refs. [15] and references therein).

Quadrupolar order has been suggested [17]; some experiments seem consistent with this picture [18], while others are less telling [19]. Experiments are consistent with two distinct energy scales in the system [19] indicating primary and secondary order parameters. A coupling or switching between the two parameters is also apparent from phenomenological arguments (see [19] for recent discussions). In all, several theories involving exotic microscopic mechanisms have been formulated and are still a matter of controversy and debate [20,21,22].

Part of the problem can be traced back to the experimental characterization of the magnetic state at low external fields. This is because the time scales from different techniques used to sample the low frequency (≤ 100 MHz) magnetic response (i.e., magnetization, NMR, µSR) do not overlap with those from techniques used to probe the spin dynamics (i.e., ND). In particular, ND measures a “static” ordered moment of about 0.02–0.03µB [23] along the c axis, which would necessarily split the ^29Si NMR line by ~80–100 Gauss (external field collinear with the spontaneous moment). This splitting is not observed [23,24], and as a result, it has been argued that the tiny moment must be fluctuating at time scales that render it invisible to NMR or even µSR probes [25].

(See, however, Ref. [14]). At first, this argument seems to suggest that NMR is unable to shed light on the nature of the phase transition. However, since NMR can sample electronic effects both directly and indirectly [20], it should also be expected to reveal other significant information about the ordered state, even if a transition to static magnetic order at T_N is not established by NMR measurements. Accordingly, the absence of line splitting might offer a chance to better characterize the transition at low fields without the added complication of static magnetic-order effects on the NMR lineshape. Of particular interest, for example, would be the possibility of observing effects due to a charge-related order parameter.

In this Letter we report a new ^29Si NMR study at low field strengths (below 6 T). We find an unambiguously, field-independent isotropic component of linewidth λ which increases below T_N to about 11.5(1.5) G, average value, at 4.2 K [26]. This component is static and
measures a distribution of local effective fields at the $^{29}$Si sites. Its temperature dependence is that of a mean-field order parameter. We argue that $\lambda$ is unrelated to the static magnetization of the sample and that, instead, it is due to a coupling of the $^{29}$Si nuclei to the "hidden order" $\lambda_{\parallel}$ in this system. We propose a coupling mechanism based on charge degrees of freedom and nuclear spin/spin interactions. We also propose further experiments to test for the nature of this coupling.

The sample used was a fine powder (particle size $\lesssim 50 \, \mu$m) embedded in Stycast 1266 epoxy and oriented in a field of 9.4 T. An alignment factor of order 90–95% was estimated for the $c$ axis orientation by measuring the magnetic susceptibility under both transverse and longitudinal external fields, and comparing it with that of a single crystal under similar conditions. The alignment method leaves a random distribution of $(a,b)$ basal-plane orientations (tetragonal structure). The $^{29}$Si spectral parameters were measured as functions of temperature and applied field for different field directions with respect to the $c$ axis.

![Signal Intensity A.U.](image)

**Fig. 1.** $^{29}$Si NMR spectra in URu$_2$Si$_2$ for $T = 14.5$ K under (a) transverse- and (b) longitudinal-field conditions. Curves: fits to Lorentzian functions of HWHM $\Gamma(H, T)$.

Spectra for $H \perp \hat{c}$ and $H \parallel \hat{c}$ [Fig. 1(a) and (b) respectively] consisted of a single narrow line. Each line could be fitted to a Lorentzian function of half-width-at-half-maximum (HWHM) $\Gamma$ for the $T$- and $H$-range reported here. We find that the linewidth can be written as $\Gamma^2(H, T) = \Gamma_m^2(H, T) + \lambda^2(T)$, where $\Gamma_m$ is the contribution due to the sample magnetization (a term proportional to $H$), and $\lambda$ is the new contribution to the width, which is zero above $T_N$.

Separating $\Gamma$ into two components is compelled by its field dependence, which we present in Fig. 2. For $H \perp \hat{c}$ and $T \sim 14.5$ K, $\Gamma$ (open circles) is weakly linear in the field magnitude above about 3–4 T and field independent below this value, with a clear non-zero intercept. The data can be fitted to the assumed form as indicated by the curve [i.e., $\lambda(14.5$ K) $\sim 7$ G]. For comparison, we present a similar study at $T = 20$ K (solid circles), where no extra width exists: the dashed curve is a straight-line fit (slope $\sim 0.6$ G/T). Similarly, the solid triangles in

![Field dependence of the full $^{29}$Si linewidth $\Gamma$.](image)

**Fig. 2.** Field dependence of the full $^{29}$Si linewidth $\Gamma$. Circles: $H \perp \hat{c}$, $T = 14.5$ K (open) and $T = 20$ K (filled). Filled triangles: $H \parallel \hat{c}$, $T = 20$ K. Open triangles: $H \parallel \hat{c}$, $T = 14.5$ K. Squares: $T = 4.2$ K. Curves: one- (dashed lines) and two- (solid) component fits to the linewidth.

Fig. 2 represent the total width for $H \parallel \hat{c}$ and $T = 20$ K. Here, the slope ($\sim 6.4$ G/T) [dashed-line fit] is more than ten times greater than for $H \perp \hat{c}$ due to the large magnetic anisotropy of the system. The open triangles and the squares are obtained for $T = 14.5$, and 4.2 K respectively. As for $H \perp \hat{c}$, the curves drawn through these data are fits to a two-component $\Gamma$. It is seen from these fits, that the orientation dependence of $\lambda$ at constant $T$ is weak or nonexistent; for example, at 14.5 K the fits are consistent with having similar intercepts, i.e., $\lambda(H \perp \hat{c}, 14.5$ K) $\sim \lambda(H \parallel \hat{c}, 14.5$ K) $\sim 7$ G. [We have also measured the orientation dependence of $\lambda$ directly; the results are discussed below.] The value of $\lambda$ at 4.2 K is clearly different, $\lambda(4.2$ K) $\sim 13(2)$ G, as one could expect for a $T$-dependent $\lambda$ (see Fig. 3 below). Finally, one can see from Fig. 3 why, in early experiments, $\lambda$ was not detected with NMR at high-$H \parallel \hat{c}$ fields [13] or for poorly aligned samples [11]. Because of the strong magnetic anisotropy in this system, the part of the width due to either the magnetization in the aligned powder, or the anisotropic residual powder pattern in poorly aligned samples, can overwhelm $\lambda$, even at low values of $H \parallel \hat{c}$.

The Knight shift $K$ has been reported previously under longitudinal and transverse field geometries [12, 13]. Since in antiferromagnetic systems one might expect a change in the orientation dependence of $K$ as $T$ crosses $T_N$ [14], we followed $K$ vs. $\theta$, the field orientation angle, above and below $T_N$. We find that the behavior of $K$ reflects only the anisotropic magnetization and does not seem to be involved in the linewidth effect we observe: the lineshape (Lorentzian-like) does not change with $\theta$ (Fig. 3) and $\lambda$ does not shift anomalously through $T_N$. This can be seen clearly in Fig. 3 (a) and (b) where we present, respectively, $K(\theta)$ and $\Gamma(\theta)$ above $(T = 20$ K; open circles) and below $(T = 4.2$ K; closed circles) $T_N$. For both temperatures, the shift can be fit to $K = \mu^2K(\theta) + (1 - \mu^2)K_\perp$, $\mu \equiv \cos \theta$ (drawn lines), as expected from crystal anisotropy [15]. The difference
in magnitude for \( \theta \to 0 \) is due to the temperature dependence of the magnetization in that direction.

The curve drawn in Fig. 3(b) for \( T = 20 \) K is expected if \( \Gamma \) represents a distribution of anisotropic shifts with two independent width components: \( \delta K_{\parallel}(T) \) and \( \delta K_{\perp} \) (i.e., \( \mu \) is not distributed, and \( \delta K_{\perp} \) is not \( T \)-dependent). We find \( \delta K_{\parallel}(20 \) K\) = 0.056\%(4) \%, and \( \delta K_{\perp} = 0.013(4) \% \). For \( T = 4.2 \) K [Fig. 3(b), closed circles], \( \lambda \) is present, and the fit is consistent with \( \lambda \) being isotropic. Here, we find \( \delta K_{\parallel}(4.2 \) K\) = 0.078\%(4) \%, and \( \lambda(4.2 \) K\) = 9.9(5) G.

Figure 4 gives the temperature dependence of \( \lambda \). The filled symbols are obtained in the transverse-field geometry by direct quadrature subtraction of the term \( \Gamma_{\text{m}}(H \perp \hat{c}) \), which is temperature independent. Since \( \Gamma_{\text{m}} \) is strongly \( T \)-dependent in the longitudinal field configuration, direct extraction of \( \lambda \) from the temperature dependence of the total width \( \Gamma \) in that geometry is not straightforward. Our procedure is as follows. We subtracted \( \Gamma_{\text{m}}(H \| \hat{c}, T) \) in this case by using the data in Fig. 2 together with a scaling of the form \( \Gamma_{\text{m}}(H \| \hat{c}, T) \propto M(H \| \hat{c}, T) \), where \( M \) is the magnetization. Random errors in the values of \( H \| \hat{c}, \theta \) and \( T \) from different experimental runs become important in deducing a good value of \( \lambda(T) \) as can be seen from the scatter of the data in this geometry. Nevertheless, the resulting values are consistent with the lack of anisotropy inferred above from field and orientation studies. The behavior of \( \lambda \) clearly signals that there exists a coupling between the \( 29\text{Si} \) nuclear spins and the electronic transition at \( T_N \). This is further corroborated by the BCS-gap-equation fit (drawn curve in Fig. 4), which indicates a mean-field-theory temperature dependence for \( \lambda \).

Of the characteristics found for \( \lambda \) during this study, its lack of strong anisotropy and its independence of the lineshift \( K \) are the hardest ones to reconcile within a simple model of magnetic order/disorder at the U sites as the origin of \( \lambda \). Although a distribution of internal fields could be produced by disorder or incommensuration in the direct (dipolar) or indirect transferred interaction between U moments and the \( 29\text{Si} \) nuclei, one would expect the linewidth to have the anisotropy of this interaction and the lineshape and width to change as a function of the external field direction. Neither of these expectations is borne out by the behavior of \( \lambda \), even though the shift \( K \) does follow the anisotropic susceptibility. Accordingly, the observed lineshape (Fig. 1) suggests that a fair number of nuclei, contributing to the center of the line, probe little or no internal field independently of the external-field orientation. Any model of magnetic disorder at the U-sites, would have to invoke a continuous distribution of moment sizes or domains that includes values all the way down to zero. Although domains of this type are found in, for example, NiO, as reported 40 years ago [18], the direction of the spontaneous moment within the domains would have to be either distributed almost isotropically (for \( \mu \sim 0.03 \mu_B \), \( 0^\circ \leq \theta \lesssim 80^\circ \)), or such that it is coupled to the direction of the applied field. In any case, such interpretation would be in striking contrast with ND and \( \mu \)SR measurements for which the correlations lie predominantly along the \( c \) axis. Thus, the behavior of \( K \) and \( \lambda \) together with ND and \( \mu \)SR results rule out static magnetic order/disorder at the U sites.

Alternatively, a qualitative explanation of the main characteristics of \( \lambda \) is achieved by invoking an indirect nuclear spin/spin interaction between unlike nuclei: \( \mathcal{H}_{ij} = A_{ij} \mathbf{I}_i \cdot \mathbf{I}_j \). For instance, \( \lambda \) is static and inhomogeneous, so it represents a distribution of time-average-effective local fields such as could be produced by \( ^{99,101} \text{Ru} \) nuclei at \( 29\text{Si} \) sites. The disorder effect would come from the fact that all isotopes involved are randomly distributed in the crystal because of their low natural abundance. This mechanism does not contribute to the line shift \( K \), which explains the independence of \( \lambda \) with respect to this parameter. The field independence of \( \lambda \) could also be explained by this mechanism; the indirect interaction is a second order effect, dominated by the excitation energy of the mediating electrons, which is in general much larger than the nuclear resonance energy.

The difficulty here is to ascertain what the unperturbed Hamiltonian ought to be in order to calculate \( A_{ij} \) first principles. For example, RKKY interactions between the nuclei [19] could be mediated either by normal conduction electrons or by renormalized

FIG. 3. Dependence of \( ^{29}\text{Si} \) Knight shift \( K \) (a) and linewidth \( \Gamma \) (b) on \( c \)-axis angle \( \theta \) in \( \text{URu}_2\text{Si}_2 \) (see text).

FIG. 4. Anomalous \( \lambda(T) \) extracted from \( ^{29}\text{Si} \) NMR linewidths in \( \text{URu}_2\text{Si}_2 \) for \( H \perp \hat{c} \) (filled circles) and \( H \| \hat{c} \) (open circles). Curve: fit to BCS-gap equation.
quasiparticles. Crude estimates based on our previous measurements of the Knight shift vs. the spin susceptibility $A_{ij} \sim 10^{-6}$ G in the former case, and $A_{ij} \sim 10^{-3}$ G in the latter. These values are negligible and would also be present in the high-temperature state. The possibility that such an effect could be amplified to $\sim 10$ G by the phase transition would require an increase of order $10^2$ in the overlap of the quasiparticle wavefunctions with the $^{29}$Si nuclei below $T_N$. Another possibility is the Suhl-Nakamura mechanism [20], in which antiferromagnetic spin waves mediate the indirect nuclear spin-spin interaction. However, in view of the fluctuating-moment picture (which is supported by our measurements) [2], the use of a static AF state for the electron system is not justified.

On the other hand, the evidence that the primary hidden order parameter $\psi$ (whatever it might be) is actually coupled to the antiferromagnetic correlations observed by ND and $\mu$SR allows one to envision the following mechanism for indirect nuclear spin interactions. Let a $^{99,101}$Ru nucleus produce a virtual excitation of the electronic state (governed by $\psi$). Because of the coupling between $\psi$ and the secondary (antiferromagnetic) order parameter $m$, a change in the electronic spin state occurs allowing the nearby $^{29}$Si nuclei to detect the interaction. For instance, if the primary order parameter is quadrupolar or that of a charge-density wave CDW, there will be an interaction between the $^{99,101}$Ru nuclei and the electric field gradients produced by the charge state of the U ions. The presence of a small external field (small compared with the excitation energy of the quadrupolar states of the electrons) produces a Zeeman splitting of the electron spin states, so that electron spin flips can occur when a quadrupolar/CDW excitation is induced by the $^{99,101}$Ru nuclei. The time average of these spin-flips would be responsible for the local change in resonance frequency at the $^{29}$Si sites. Unfortunately, we lack at present a good characterization of the electronic ground state to proceed further in this direction. Nevertheless, we suggest that our conjecture be checked theoretically using the different microscopic models that have been proposed in the literature.

In conclusion, magnetic U-site order/disorder as the origin of the anomalous linewidth $\lambda$ can be ruled out by combined NMR and ND/$\mu$SR experiments. Furthermore, we discovered a relationship between $^{29}$Si NMR linewidth and the primary “hidden” order parameter of the 17 K transition. Double resonance experiments in which the nuclei are decoupled by an rf field and NMR in isotopically enriched samples need to be performed as functions of $T$ and $H$ in order to elucidate the extent to which indirect coupling between unlike nuclei is the source of the NMR broadening. Finally, we point out that we appear to be observing the effects of charge degrees of freedom in URu$_2$Si$_2$ by means of $^{29}$Si, which has a spin 1/2 and cannot sample quadrupolar effects directly. The proposed experiments should further test this conjecture.

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