Detection and decoherence of level-crossing resonances of ^{8}Li in Cu

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Level-crossing resonances are observed for spin-polarized ^{8}Li in copper at 200 K. The positions of the resonances as a function of magnetic field and crystal orientation are a precise measure of the induced quadrupolar interaction on the nearest-neighbor Cu spins and provide unambiguous evidence that ^{8}Li occupies a substitutional site. The resonances are detected as enhancements in the ^{8}Li spin relaxation rate and are much broader than predicted from a static spin Hamiltonian. A strong collision model is used to extract a decoherence time as a result of the dipolar coupling of the ^{8}Li-Cu subsystem to the surrounding nuclear spin bath.

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Level-crossing resonance is a powerful method for characterizing the local electronic structure of the positive muon or a radioactive nucleus implanted into a crystalline solid.[1–5] In general, one expects to observe a coherent oscillation in the spin polarization on resonance as a result of mixing of two nearly degenerate spin states.[6] However, any such quantum mechanical process will lose coherence when placed in contact with a fluctuating environment. Such decoherence has broad implications. For example, it is central to many problems related to quantum computing, e.g., the storage and manipulation of quantum information using q-bits.[7–10] In particular, in studies of magnetic molecules, the nuclear spin bath disrupts coherent oscillations of the magnetization at level crossings and leads to incoherent tunneling relaxation.[11]

In this Brief Report, we report observations of level-crossing resonances associated with dilute ^{8}Li in single crystal Cu as a function of orientation and magnetic field, using β-detected nuclear magnetic resonance (β-NMR).[12] The subsystem in this case is ^{8}Li along with the shell of nearest-neighbor Cu nuclei which experience an electric field gradient (EFG) caused by the ^{8}Li. The EFG's strength diminishes rapidly with distance from the ^{8}Li and hence partly decouples the nearest-neighbor Cu spins from more distant neighbors and results in a kind of defect "molecule," which we denote as ^{8}Li-Cu. Level-crossing resonances are anticipated to occur at magnetic fields where the Larmor splitting of the ^{8}Li is matched to a Cu nuclear spin splitting that depends on both the Zeeman and induced quadrupolar interactions. The magnetic dipolar coupling between the ^{8}Li and Cu nuclei lifts the degeneracy between the two levels, giving rise to a coherent oscillation in the ^{8}Li spin polarization in the absence of all other spins. The positions of such resonances as a function of magnetic field and crystal orientation are an unambiguous signature of the defect molecule and provide detailed structural information. The current study establishes that the site of implanted ^{8}Li can be determined via its level crossing resonances and therefore has important applications in experiments where the ^{8}Li is being used as a local probe of bulk materials, ultrathin films, and interfaces.[13–22] The resonances are detectable as an enhancement in the ^{8}Li $1/T_1$ due to cross relaxation with the neighboring Cu spins. The large width of the resonances is attributed to fluctuations in the surrounding nuclear spin bath, which are treated using a simple strong collision model.

Although level-crossing resonances have been used to characterize the electronic structure of both muons and radioactive nuclei,[1–5] there are few measurements on ^{8}Li that have a relatively long radioactive lifetime (1.2 s) compared with the μ^+ or other β-NMR isotopes. To our knowledge, the only existing measurement is in neutron activated $LiNbO_3$ where ^{8}Li remains at the host Li site.[23] Cu metal is a good candidate for observing level-crossing resonances of implanted ^{8}Li because of its simple face-centered-cubic (fcc) crystal structure and suitable nuclear spin properties. Each Cu nucleus has a spin of 3/2 and hence a quadrupole moment; there are two isotopes ^{63}Cu and ^{65}Cu with 69.17% and 30.83% natural abundance, respectively. Previous β-NMR studies of the site-dependent Knight shifts of ^{8}Li in fcc elemental metals[13,14,24] indicate a significant fraction of ^{8}Li occupies a cubic interstitial (I) site at low T and a conversion to a different cubic site occurs at higher T. The higher T site is thought to be substitutional (S) but, so far, there has been no concrete evidence to prove this. In Cu, the $I \rightarrow S$ conversion takes place at $\sim$ 150 K.[14] However, this conflicts with earlier studies of the ^{8}Li β-NMR linewidth, which suggest that the S site is occupied at 100 K.[25]

The β-NMR experiment was performed at the Isotope Separator and Accelerator (ISAC) depth-resolved β-NMR facility[12,13] at TRIUMF in Vancouver, Canada using a highly spin-polarized ^{8}Li$^+$ beam.[26] The beam is implanted at a rate of $\sim 10^6$ s into the sample mounted on a cold-finger cryostat under ultrahigh vacuum (UHV). Level crossings were detected by measuring the time dependence of the ^{8}Li polarization

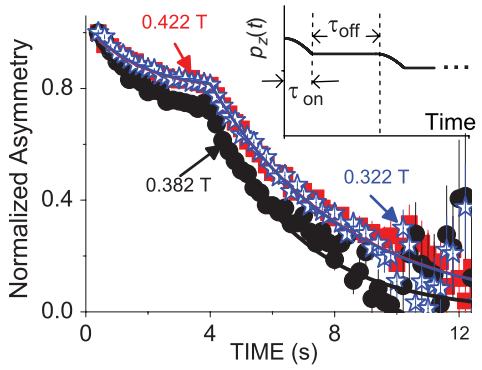

FIG. 1. (Color online) Representative examples of the normalized ^{8}Li asymmetry at 200 K in Cu with **B** parallel to a ⟨110⟩ crystalline direction. The solid lines are fits to the data. The inset illustrates the time evolution of the ^{8}Li polarization in the strong collision model of the relaxation rate.

as a function of the applied magnetic field. In particular, we measured the beta decay asymmetry using appropriately positioned scintillation counters; this enables us to extract the ^{8}Li polarization since an electron is emitted preferentially opposite to the direction of the polarization at the time of the decay.[27] All measurements were carried out using a beam-on period of 4 s followed by 8 s of beam-off. Details on the experimental setup are given elsewhere.[15] Three 99.999% purity Cu single crystals (Accumet Materials Company) of dimensions 8 mm × 10 mm × 1 mm, with ⟨111⟩, ⟨110⟩, and ⟨100⟩ directions perpendicular to their faces, were studied. Prior to the experiment, the polished surface of each sample was first prepared by sputter cleaning, UHV annealed, and capped with a thin Au layer (∼ 4 nm) to prevent surface oxidation. In the β-NMR study, the ^{8}Li$^+$ was implanted at 28 keV, and hence has an average penetration depth of ∼ 88 nm and a straggling of ∼ 41 nm (as calculated using TRIM.SP[28]). The initial ^{8}Li spin-polarization, the normal to the crystal face, and **B** are parallel. All measurements were carried out at 200 K, which is well above where the $I \rightarrow S$ site transition is thought to occur.

Figure 1 shows examples of the normalized beta decay asymmetry as a function of time, which equals the spin-polarization function $p_z(t)$ convoluted with a square beam pulse.[15] The data in all three samples and at all fields were fitted well by assuming that the time decay of the ^{8}Li polarization is a single exponential with decay rate $1/T_1$. (The kink at 4 s is due to the turn-off of the ^{8}Li$^+$ beam—see Ref. 15). It is clear from Fig. 1 that $1/T_1$ is enhanced at 0.382 T compared to the other two fields shown. The full field dependences of $1/T_1$ at 200 K for **B** parallel to the three major crystalline directions (i.e., the three samples) are shown in the upper three panels of Fig. 2. Note at high fields, particularly obvious in the ⟨111⟩ and ⟨100⟩ orientations, $1/T_1$ is field independent with a value of ~ 0.225 s^{-1}. This field independent term is attributed to the well-known Korringa relaxation, i.e., a consequence of the Fermi contact interaction of the ^{8}Li and resultant scattering with the conduction electrons at the Fermi surface.[14] There is also an overall increase in $1/T_1$ as **B** is decreased toward zero. This is due to many unresolved level crossings near zero field[14,19] and does not provide spectroscopic information on the ^{8}Li. The interest in this paper lies in the pronounced enhancements in $1/T_1$ at specific magnetic fields, which are attributed to resolved level crossings in the ^{8}Li-Cu subsystem.

In order to understand the level-crossing resonances shown in Fig. 2, we first introduce a static spin Hamiltonian to describe ^{8}Li located at a cubic site:

$$\begin{aligned}\mathcal{H} = &-\tilde{\gamma}_{\mathrm{Li}} B I_z - \tilde{\gamma}_{\mathrm{Cu}} B K_z \\ &+ q_{\mathrm{Cu}}\left[3K_{z'}^2 - K^2 + \eta\left(K_{x'}^2 - K_{y'}^2\right)\right] \\ &+ D(I_{x'}K_{x'} + I_{y'}K_{y'} - 2I_{z'}K_{z'}),\end{aligned} \tag{1}$$

where **I** and **K** are the ^{8}Li and the Cu spin operators, respectively. This model involves only one nucleus but is sufficient to address the positions of the resonances. The first two terms in Eq. (1) represent the Zeeman interactions of ^{8}Li and the Cu, with the assumption that **B** is applied along the z axis. Here, $\tilde{\gamma}_{\mathrm{Li}} = 6.3015$ MHz/T is the gyromagnetic ratio of the ^{8}Li nucleus. The gyromagnetic ratio $\tilde{\gamma}_{\mathrm{Cu}}$ of the ^{63}Cu and ^{65}Cu nuclei are $^{63}\tilde{\gamma} = 11.319$ MHz/T and $^{65}\tilde{\gamma} = 12.103$ MHz/T. The third term in Eq. (1) describes the EFG due to the ^{8}Li on the Cu nucleus. q_{Cu} is the quadrupole coupling constant of the Cu nucleus. For ^{8}Li at a S site, the EFG induced on the Cu site is likely nonaxial and therefore the dimensionless asymmetry parameter $\eta = (V_{x'x'} - V_{y'y'})/V_{z'z'}$ is needed, where $V_{x'x'}$, $V_{y'y'}$, and $V_{z'z'}$ are the principal EFG components (with $|V_{x'x'}| \leqslant |V_{y'y'}| \leqslant |V_{z'z'}|$). The x', y', and z' (and x, y, and z) directions are illustrated for a representative configuration in Fig. 2; cf. Ref. 29. Note that $q_{\mathrm{Cu}} = \frac{eQ(\mathrm{Cu})V_{z'z'}}{h4K(2K-1)}$, where the nuclear quadrupole moments for the two isotopes[30,31] are $Q(^{63}\mathrm{Cu}) = -0.211$ barn and $Q(^{65}\mathrm{Cu}) = -0.195$ barn. Finally, the dipolar tensor, i.e., fourth term in Eq. (1), is assumed to be axially symmetric about z'. The dipolar constant is $D = \mu_0 h\tilde{\gamma}_{\mathrm{Cu}}\tilde{\gamma}_{\mathrm{Li}}/4\pi r^3$, where r is the ^{8}Li-Cu separation. There are no terms in Eq. (1) that can produce $1/T_1$ relaxation (see below).

The time dependent ^{8}Li nuclear spin polarization along **B** can be calculated from Eq. (1) via

$$\begin{aligned}p_z(t) &= \frac{\mathrm{Tr}[I_z e^{-i\mathcal{H}t/\hbar}\rho(0)e^{i\mathcal{H}t/\hbar}]}{\mathrm{Tr}[I_z\rho(0)]} \\ &= \left(1 - \sum_j a_j\right) + \sum_j a_j \cos\omega_j t,\end{aligned} \tag{2}$$

where $\rho(0)$ is the density matrix describing the initial ^{8}Li spin state, while a_j and ω_j are the amplitudes and precession frequencies that can be calculated from the eigenvectors and eigenvalues of $\mathcal{H}$, respectively. We assume in our calculations that the ^{8}Li is initially fully polarized and the Cu nuclei are unpolarized. Note, in general, from Eq. (2), $p_z(t)$ consists of a sum of coherent oscillations. In a high magnetic field, away from any level crossings, the amplitudes for all frequencies are nearly zero and the calculated ^{8}Li polarization is time independent. On a resonance two spin states of the combined ^{8}Li-Cu subsystem are mixed by the magnetic dipolar interaction, leading to a coherent oscillation in $p_z(t)$ with a frequency on the order of D, i.e., kHz.

Approaches based on analogous versions of Eq. (1) and Eq. (2) have been applied very successfully to describing the level crossing resonances of other radioactive spin-polarized

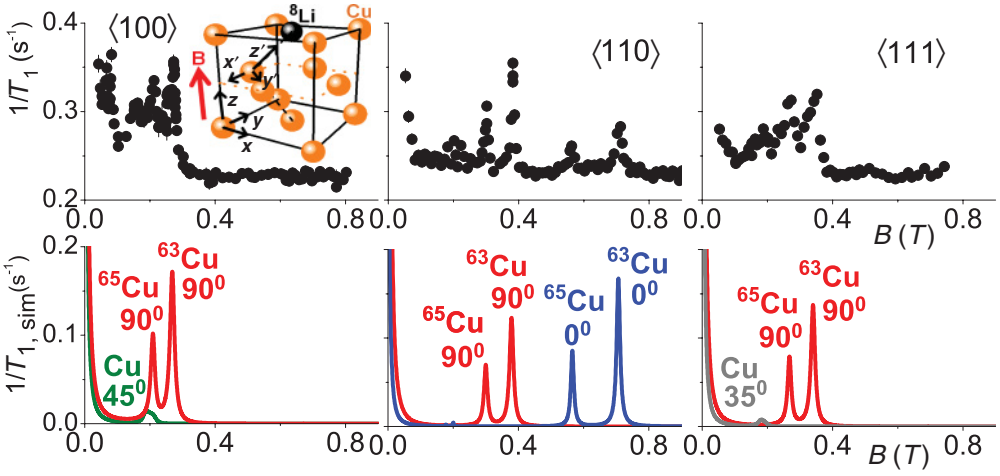

FIG. 2. (Color online) Top three panels are the experimental $1/T_1$ for **B** applied parallel to the ⟨100⟩, ⟨110⟩, and ⟨111⟩ orientations of Cu. The lower three panels are simulations as described in the text. Also shown is the fcc lattice of Cu with the ^{8}Li in a substitutional site.

probes such as the μ^+.[4–6] However, they cannot explain the ^{8}Li resonances shown in Fig. 2. For example, at a level crossing resonance, Eq. (1) and Eq. (2) predict a drop in the initial asymmetry equal to the amplitude of the oscillation ($\sim 10\%$) since the on-resonance frequencies are too high to resolve experimentally.[32] Instead we observe a small enhancement in the spin relaxation rate with no clear change in the asymmetry. The observation that the resonances manifest as enhancements in $1/T_1$ relaxation implies that the approach described by Eq. (1) and Eq. (2), based on the static Hamiltonian, is not adequate. Furthermore, the intrinsic width of a resonance, i.e., the field dependence of the on-resonance a_j, should only be a few gauss [calculated from Eq. (2)], whereas our experimental resonances are about 100 times wider. We attribute these differences to the surrounding nuclear spins, whose effect is amplified in the case of ^{8}Li compared to the μ^+.

In particular, the magnetic dipolar interaction between ^{8}Li and its nearest neighbor Cu is comparable to the Cu-Cu dipolar interaction, whereas in the case of the muon the muon-dipolar interaction dominates. The ^{8}Li-Cu subsystem has a large number of nearest neighbors and a spin fluctuation in any one of these spins can disrupt the coherent transfer of polarization in ^{8}Li-Cu. Together these imply that the decoherence rate is much greater than the oscillation frequency on resonance, which is typically not the situation in the case of a muon.

We apply a strong-collision model[33,34] in order to account for fluctuations in the nuclear spin bath and to model the ^{8}Li $1/T_1$. Here, "strong collision" refers to a fluctuation in one of the neighboring nuclear spins which may arise, for example, from T_2 processes with other Cu nuclei. This disrupts the coherent oscillation of ^{8}Li-Cu. Consider a ^{8}Li-Cu which starts off at $t = 0$ on resonance. The polarization evolves coherently for a mean time τ_{on} before a "collision."Collisions take the ^{8}Li-Cu off resonance for a mean time τ_{off}, during which the ^{8}Li polarization is time independent. After a number of fluctuations the ^{8}Li will come back on resonance. The central assumption of the model is that upon return to resonance, the density matrix is reset to its initial value, except with a slightly reduced ^{8}Li polarization and also with a different phase. i.e., the subsystem continues to evolve as at $t = 0$ but with a reduced initial amplitude. The cycle is illustrated in the inset of Fig. 1. Under these assumptions, $1/T_1$ is the product of two quantities[34]: (i) the time-averaged fractional loss of (the oscillatory components of) the polarization after each cycle, i.e., $\langle 1 - p_z(t)\rangle$, and (ii) the cycle rate, i.e., $1/(\tau_{\mathrm{on}} + \tau_{\mathrm{off}})$. Hence

$$1/T_1 = \left(\frac{1}{\tau_{\mathrm{on}} + \tau_{\mathrm{off}}}\right)\left(1 - \frac{1}{\tau_{\mathrm{on}}}\int_0^\infty e^{-\frac{t}{\tau_{\mathrm{on}}}} p_z(t)dt\right)$$
$$= \left(\frac{1}{\tau_{\mathrm{on}} + \tau_{\mathrm{off}}}\right)\sum_j a_j \frac{\omega_j^2}{\left(\frac{1}{\tau_{\mathrm{on}}}\right)^2 + \omega_j^2}, \qquad (3)$$

where a_j and ω_j are the same as in Eq. (2). Note the amount of polarization loss per cycle only depends on τ_{on} since $p_z(t)$ is time independent during τ_{off}.

The positions of level-crossing resonances shown in Fig. 2 provide an unambiguous identification of the ^{8}Li location in the crystal. In particular, the site problem is very much overdetermined since once the site has been correctly assigned there are only two adjustable parameters [q_{Cu} (for one isotope) and η].[35] Inserting these into Eq. (2) [or Eq. (3)] must reproduce the positions of all resonances from the nearest neighbors in the three orientations. This is indeed the case when we assume that ^{8}Li is in a S site, as indicated by the comparisons between the experimental data in the top three panels of Fig. 2 with the simulations shown in the bottom three panels. These simulations are carried out assuming interactions with one Cu nucleus with the appropriate ^{63}Cu and ^{65}Cu isotopic abundances. The relevant values are $q(^{63}\mathrm{Cu}) = 1.0806 \times q(^{65}\mathrm{Cu}) = 0.591(4)$ MHz and $\eta = 0.136(8)$. The angle θ in Fig. 2 denotes the angle between **B** and the direction between ^{8}Li and a nearest-neighbor Cu nucleus, i.e., the angle between z and z'. (In an undistorted lattice, the θ's are known once the site is assumed.[36]) The close agreement between the simulated and experimental resonance positions establishes firmly that ^{8}Li is located in the S site at 200 K. This is strong confirmation of recent work on fcc metals that suggests the high T site is S,[13,14,24] and suggests that the ^{8}Li site transition

around 100–150 K seen in many fcc metals is $I \rightarrow S$. This in turn implies the low T site in Cu is I and *not* S, as indicated by earlier work.[25]

We now discuss the amplitudes and widths of the resonances in Fig. 2. The simulations shown in the lower panels of Fig. 2 were carried out assuming a single nearest Cu neighbor at a particular value of θ, as well as an undistorted lattice with nearest neighbor distance between ^{8}Li and Cu of 2.56 Å.[37] Then, Eq. (3) is used to calculate the $1/T_1$; note that τ_{on} modifies the widths of the peaks, while the sum $\tau_{on} + \tau_{off}$ modifies the amplitude. Reasonable reproduction of the data requires $\tau_{on} \sim 5$ μs and $\tau_{off} \sim 250$ μs. Note that τ_{on} is considerably shorter than the Cu-Cu spin-spin relaxation time[38] $T_2 \sim 100$ μs. The difference is reasonable after one considers that the substitutional ^{8}Li Cu pair has nearly 20 nearest neighbors, and the number will increase with the number of Cu nuclei on-resonance. A fluctuation in any one of these neighbors has a chance to disrupt the coherence. The τ_{off} is much longer than τ_{on}, implying that many fluctuations are needed to "restart" the cycle and that the ^{8}Li-Cu spends a significant fraction of time off-resonance. This is also understandable given the narrowness of the intrinsic resonance (i.e., without coupling to the spin bath), which reduces the chance that a fluctuation of the spin bath will return the ^{8}Li-Cu back on resonance. The present results, along with the strong collision model, are relevant to any coherent process where the coupling to the bath is appreciable, i.e., the coherent frequency or tunnel splitting of the subsystem is small compared the bath's fluctuation rate. Finally, we note that the nuclear spin dynamics could in principle be simulated using a general Louiville formalism, which has been used to account for the effect of diffusion on the level-crossing resonances of ^{12}B in Cu.[39] However, the strong collision model, as formulated here, explicitly takes into account the on- and off-resonance cycling expected at such level crossings. Furthermore, the strong collision model embodied in Eq. (3) is more intuitive and much easier to apply.

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[35] Reasonable dipolar interactions are on the order of kHz, while the quadrupole interactions are in the range of MHz. Hence the positions of the resonances are very *in*sensitive to the dipolar interaction.
[36] ^{8}Li in a S site has 12 Cu nearest neighbors. When $\mathbf{B} \parallel \langle 100 \rangle$, there are eight equivalent nuclei at $\theta = 45^\circ$ and four neighbors at $\theta = 90^\circ$. When $\mathbf{B} \parallel \langle 110 \rangle$, there are two equivalent Cu nuclei at 0°,

eight nuclei at 60°, and two nuclei at 90°. When **B** ∥ ⟨111⟩, there are six equivalent Cu nuclei at 35.3° and six nuclei at 90°.

[37]The dipolar coupling constants $^{63}D = \mu_0 h^{63}\tilde{\gamma}\tilde{\gamma}_{\mathrm{Li}}/4\pi r^3 = 283$ Hz and $^{65}D = 303$ Hz in the undistorted Cu lattice.

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