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Phys. Rev. B **90**, 165127 — Published 21 October 2014

DOI: [10.1103/PhysRevB.90.165127](https://doi.org/10.1103/PhysRevB.90.165127)

# NMR relaxation in the Topological Kondo Insulator SmB<sub>6</sub>

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(Dated: October 1, 2014)

SmB<sub>6</sub> has been predicted to be a strong topological Kondo insulator and experimentally it has been confirmed that at low temperatures the electrical conductivity only takes place at the surfaces of the crystal. We study the temperature and magnetic field dependence of the NMR Knight shift and relaxation rate arising from the topological conduction states. For the clean surface the Landau quantization of the surface states gives rise to highly degenerate discrete levels for which the Knight shift is proportional to the magnetic field  $B$  and inversely proportional to the temperature  $T$ . The relaxation rate,  $1/T_1$ , is not Korringa-like. For the more realistic case of a surface with a low concentration of defects (dirty limit) the scattering of the electrons leads to a broadening of the Landau levels and hence to a finite density of states. The mildly dirty surface case leads to a  $T$ -independent Knight shift proportional to  $B$  and a Korringa-like  $1/T_1$  at low  $T$ . The wave functions of the surface states are expected to fall off exponentially with distance from the surface giving rise to a superposition of relaxation times, i.e. a stretched exponential. It is questionable that the experimental <sup>11</sup>B Knight shift and relaxation rate arise from the surface states of the TKI. An alternative explanation is that the bulk susceptibility and the <sup>11</sup>B NMR properties are the consequence of the in-gap bulk states originating from magnetic exciton bound states proposed by Riseborough [Phys. Rev. B **68**, 235213 (2003)].

PACS numbers: 71.27.+a, 74.50.+r, 72.15.Qm, 75.20.Hr

## I. INTRODUCTION

Kondo insulators are stoichiometric compounds with small-gap semiconducting properties.<sup>1,2</sup> Most are non-magnetic with a Van Vleck-like low-temperature susceptibility and a low- $T$  resistivity and electronic specific heat following an exponential activation law consistent with a gap in the density of states. Kondo insulators are not perfect semiconductors, because the gap is frequently only a pseudogap and/or there are intrinsic in-gap states with a large magnetic response.

SmB<sub>6</sub> is a Kondo insulator with a small gap originating from the hybridization between a narrow  $4f$  band and broad conduction bands. At ambient pressure, SmB<sub>6</sub> is a homogeneously intermediate valence material of valence  $\sim 2.7$  with a ratio of the  $4f^6$  to  $4f^55d$  configurations of about 3:7.<sup>3,4</sup> The indirect gap, determined from the resistivity, is approximately 54 K.<sup>5</sup> There is evidence for intrinsic “in-gap” bound states from the  $T$  dependence of the optical transmission and reflectivity through films,<sup>6,7</sup> Raman scattering,<sup>8</sup> neutron scattering experiments,<sup>9,10</sup> low- $T$  specific heat,<sup>11</sup> susceptibility<sup>12</sup> and NMR.<sup>13–15</sup> In earlier days the “in-gap” states have been attributed to magnetic excitations (exciton-like bound states) due to AF correlations.<sup>16</sup> The “in-gap” states are very sensitive to an external magnetic field.<sup>15</sup> Remarkable is also the saturation of the resistivity below 4K as the temperature is reduced, indicating that at very low  $T$  SmB<sub>6</sub> is not compatible with the picture of a semiconductor/insulator. The “in-gap” states in the susceptibility emerge at a higher temperature ( $\sim 20$  K) than the temperature at which the low  $T$  resistivity saturates. The gap of SmB<sub>6</sub> is also extremely sensitive to pressure.<sup>5,17</sup>

Time-reversal invariant band insulators can be classi-

fied by the topological structure of their ground state wave function.<sup>18–21</sup> In these “topological insulators” a strong spin-orbit interaction leads to a ground state that is topologically distinct from vacuum and gives rise to gapless surface excitations. Examples for quantum spin Hall insulators are graphene and HgTe/CdTe quantum well structures, and for 3D topological insulators Bi<sub>1– $x$</sub> Sb <sub>$x$</sub> , Bi<sub>2</sub>Se<sub>3</sub> and Bi<sub>2</sub>Te<sub>3</sub> (see Ref. 21). Dzero *et al.*<sup>22</sup> showed that Kondo insulators can also be topologically classified. Kramers ions with  $4f$ -states have a naturally strong spin-orbit coupling and are states with odd parity under inversion. Through the hybridization the strong spin-orbit interaction is embedded as well into the conduction band. SmB<sub>6</sub> has a simple cubic structure and been postulated as a candidate for a strong topological Kondo insulator (TKI)<sup>22–25</sup> (see also Refs. 26–28). For Kramers doublet ions intermediate valence is required for a strong TKI.<sup>22,23</sup> According to Raman scattering measurements,<sup>8</sup> specific heat data,<sup>11</sup> band structure calculations<sup>24</sup> and the analogy to CeB<sub>6</sub>, the ground state of bulk SmB<sub>6</sub> is a  $\Gamma_8$  quartet, which further favors a strong TKI.<sup>24,29</sup> The symmetry for the surface states is tetragonal, so that the surface ground state corresponds to a doublet (two Kramers states built from the  $\Gamma_8$  quartet). SmB<sub>6</sub> is a stoichiometric insulator and samples of high purity have been grown.

It has been proposed that the low  $T$  plateau in the resistivity of SmB<sub>6</sub> arises from the topological surface states. This has been experimentally verified by Wolgast *et al.*<sup>30</sup> who designed a novel contact configuration for a thin film-like sample and were able to distinguish bulk-dominated conduction from surface-dominated conduction. Below 4 K the bulk conductivity is frozen out and only the surface conduction remains. This shows that the low  $T$  plateau of the resistivity is due to surface con-

duction with a fully insulating bulk. Further evidence for surface conductivity has been reported in Refs. 31 and 32, where the electric conductivity and the Hall effect are proven thickness-independent, and magnetic and non-magnetic doping results in contrasting behaviors.<sup>32</sup> The effects of Kondo holes and non-magnetic impurities on the TKI states has been investigated theoretically in Ref. 33.

The topology of the Fermi surface of the surface states has been mapped using torque magnetometry.<sup>34</sup> The observed Fermi surface suggests two pockets of 2D surface states on the (101)-plane denoted with  $\alpha$  and  $\beta$ , respectively. The  $\alpha$ -pocket displays the characteristic angular dependence of a cylindrical 2D band, while the angular dependence of the  $\beta$ -pocket is flat. The masses are light and the Dingle temperatures rather large.<sup>34</sup> Furthermore, extrapolating the Landau levels to the infinite magnetic field limit leads to an intercept of  $-1/2$  for both pockets, which is characteristic of 2D electronic systems. The surface electronic structure was also probed using laser-based ARPES.<sup>35,36</sup> At low  $T$  the low-lying states form electron-like Fermi surface pockets enclosing the X (possibly the  $\alpha$ -pocket) and  $\Gamma$  (possibly the  $\beta$ -pocket) points of the surface Brillouin zone and disappear above 15 K.<sup>35</sup> Angular-dependent magnetoresistance measurements reveal periodicities of  $90^\circ$  at 5 K and  $180^\circ$  at low temperature<sup>37,38</sup> and that the surface states are dependent on the surface roughness.<sup>37</sup> Weak antilocalization and a linear magnetoresistance was observed in Ref. 39. Further ARPES and scanning tunneling microscopy studies of the insulating gap of  $\text{SmB}_6$  and surface states can be found in Refs. 40 and 41. The surface of  $\text{SmB}_6$  has also been studied via point-contact spectroscopy.<sup>42</sup>

The question of the exact topological nature of the surface states of  $\text{SmB}_6$  is still open, in particular a definite confirmation of the chirality of these states is still missing.<sup>32</sup> Furthermore, the question: Are the topological surface states the magnetic “in-gap” states mentioned above as observed in the susceptibility and the  $^{11}\text{B}$  NMR relaxation?<sup>15</sup> still needs an answer. It has been suggested by Takimoto<sup>24</sup> that since the magnetic field breaks the time-reversal symmetry, the surface states are gapped and the in-gap states are just the metallic surface states of the topological insulator. In this paper we investigate the possibility of a Korringa-like relaxation of the  $^{11}\text{B}$  nuclei arising from the topological surface states in  $\text{SmB}_6$ . This is directly related to the in-gap states.

In the temperature range of  $20 \leq T \leq 100$  K, the relaxation rate is field-independent and decreases down to  $1/20$  of that at 100 K with decreasing temperature, as a consequence of the opening of the indirect semiconductor Kondo gap.<sup>15</sup> The relaxation rate is then Korringa-like with the additional temperature dependence of the density of states. In this temperature range  $1/T_1$  then follows the electronic bulk properties. Below 20 K, however,  $1/T_1$  acquires a marked field-dependence. With increasing magnetic field the relaxation rate is suppressed further leading to long relaxation times, which was at-

tributed to the suppression of the “in-gap” states.<sup>15</sup> Hence, it is apparent that there are two different mechanisms, namely the standard Korringa relaxation for  $T > 20$  K and a relaxation with the in-gap states at lower  $T$ . There are several open questions: Is this suppression of the relaxation at low  $T$  due to the Landau quantization of the topological surface states? Since the wave functions of the surface states are expected to fall off exponentially with distance from the surface, the  $^{11}\text{B}$  nuclei should have different  $T_1$  depending on their distance from the surface. Hence, is the relaxation a superposition of many exponentials, i.e. a “stretched” exponential? Are there other states that do not contribute to the electrical conductivity responsible for the NMR relaxation, such as the magnetic excitons proposed by Riseborough?<sup>16</sup>

The remainder of the paper is organized as follows. In Sect. II we define the model Hamiltonian interpolating between the Dirac cone and a parabolic dispersion as a function of the Fermi level. The energies of the Landau quantized states then changes with  $N$  from a  $\sqrt{BN}$ -dependence to the standard  $B(N + 1/2)$ -dependence, where  $N$  is the quantum number of the Landau level. The latter is the dependence observed in the quantum oscillations of the magnetization.<sup>34</sup> In Sect. III we introduce the exchange interaction Hamiltonian between the Landau quantized surface conduction states and the nuclear spin states. The Knight shift and the relaxation rate are calculated for both, a pure strong TKI and the dirty limit, where nonmagnetic defects broaden the Landau levels without breaking the time-reversal symmetry. Conclusions follow in Section IV.

## II. MODEL

The band structure for  $\text{SmB}_6$  seems to indicate that the surfaces have three Dirac cones, one at the  $\bar{\Gamma}$ -point and the other two at the  $\bar{X}(\bar{Y})$ -points of the surface Brillouin zone.<sup>24</sup> To simplify, we work with only one band and address the superposition of more bands in the Conclusions section. We consider the following modified 2D Dirac Hamiltonian

$$\mathcal{H}_0 = v_F(\sigma_x p_x + \sigma_y p_y) + \frac{1}{2m}\sigma_z(p_x^2 + p_y^2), \quad (1)$$

where  $v_F$  is the Fermi velocity,  $m$  is an effective mass and  $\sigma_i$  represent the Pauli matrices. The first term in Eq. (1) corresponds to a Dirac cone, while the second term represents a standard parabolic dispersion (Schrödinger limit). In the absence of a magnetic field the wave functions are plane waves,  $\psi(x, y, t) = u \exp[i(k_x x + k_y y) - iEt]$ , where  $u$  is a spinor, and the energy is given by

$$E^2 = (v_F k)^2 + (k^2/2m)^2, \quad (2)$$

where  $k = \sqrt{k_x^2 + k_y^2}$ . For small  $k$  the dispersion is then linear,  $E = v_F k$ , while for large  $k$  the second term dominates and the dispersion is parabolic,  $E = k^2/2m$ .

The semiclassical cyclotron mass is defined as the derivative of the cross section  $A$  of the Fermi surface with respect to the energy

$$m^* = (\partial A / \partial E) / (2\pi) = \frac{E}{v_F^2} \left( 1 + \frac{E^2}{m^2 v_F^4} \right)^{-1/2}, \quad (3)$$

which interpolates between the Dirac limit at low  $E$ ,  $m^* = E/v_F^2$  and Schrödinger case,  $m^* = m$ , for large  $E$ . The Onsager quantization rules (Bohr-Sommerfeld quantization) are obtained from  $A_N = (2\pi e/c)B(N + \gamma)$ , where  $e$  is the electron charge and  $\gamma$  is a constant less than  $1/2$ , leading to  $E_N = v_F \sqrt{(2e/c)BN}$  in the Dirac limit ( $\gamma \rightarrow 0$ ) and to  $E_N = (e/mc)B(N + 1/2)$  in the Schrödinger limit ( $\gamma = 1/2$ ). The crossover energy between the Dirac and Schrödinger regimes is approximately  $E_c = mv_F^2$ , which for  $m = 0.1m_e$  ( $m_e$  is the free electron mass) and  $v_F = 6 \times 10^7$  cm/s (from Ref. (34)) yields  $E_c = 0.2$  eV.

The quantum mechanical solution in the presence of a magnetic field is obtained using the minimal substitution  $\mathbf{p} \rightarrow \mathbf{p} + (e/c)\mathbf{A}$  in Eq. (1). Choosing the asymmetric Landau gauge,  $\mathbf{A} = B(-y, 0)$ , the wave function has the form  $\psi(x, y, t) = u \exp(ik_x x - iEt)$ , where we denote with  $\phi_1(y)$  and  $\phi_2(y)$  the two components of the spinor  $u$ . Eq. (1) then leads to two coupled differential equations for  $\phi_1(y)$  and  $\phi_2(y)$ <sup>43</sup>

$$\begin{aligned} v_F \left[ (k_x - m\omega y) - ip_y \right] \phi_2(y) + \\ \frac{1}{2m} \left[ (k_x - m\omega y)^2 + p_y^2 \right] \phi_1(y) &= E \phi_1(y), \\ v_F \left[ (k_x - m\omega y) + ip_y \right] \phi_1(y) - \\ \frac{1}{2m} \left[ (k_x - m\omega y)^2 + p_y^2 \right] \phi_2(y) &= E \phi_2(y) \end{aligned} \quad (4)$$

where  $\omega = eB/mc$ . Denoting  $y_0 = k_x/m\omega$  and  $\bar{y} = y - y_0$  the solution of the coupled equations is

$$\begin{aligned} \phi_1(y) &= \varphi_{N-1}(\bar{y}), \quad \phi_2(y) = \varphi_N(\bar{y}), \\ \left( E_N + \frac{\omega}{2} \right)^2 &= \omega^2 N^2 + 2m\omega v_F^2 N, \end{aligned} \quad (5)$$

where  $\varphi_N(y)$  is the normalized harmonic oscillator wave function of frequency  $\omega$  and quantum number  $N$ . The expression for the energy contains the Dirac ( $\omega$  is small) and Schrödinger (neglecting the last term) cases as special limits. Dividing the expression for the energy by  $B^2$  and taking the limit  $B \rightarrow \infty$  (extreme quantum limit, i.e.  $N = 0$ ) we obtain that  $E \rightarrow -\omega/2$ , which is in agreement with the corresponding extrapolation of the quantum oscillations in the torque magnetometry experiment.<sup>34</sup> These levels were found to be equally spaced<sup>34</sup> as expected for the Schrödinger limit. The position of the Landau levels for a field of 10 T is shown in Fig. 1(a).

The Landau levels are highly degenerate. Using periodic boundary conditions for a linear dimension  $L$  of the sample we have  $k_x = 2\pi n_x/L$ , where  $n_x$  is a positive integer. The equilibrium position of the oscillators,  $y_0$ , has

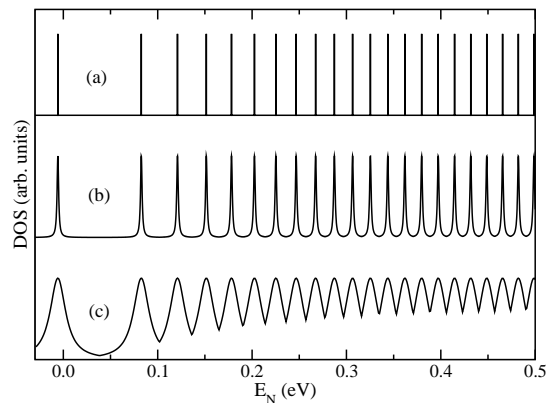


FIG. 1: Density of states in arbitrary units as a function of the Landau level positions according to Eq. (5) for the parameters discussed in the text. (a) Delta functions in the clean limit, (b) slightly broadened levels due to scattering off defects, and (c) DOS in the dirty limit. In (b) and (c) the sharp Landau levels were replaced by a Lorentzian lineshape, i.e. a constant imaginary part of the selfenergy, although the real selfenergy is energy dependent. Note that the defects introduce a background DOS. The shifts of the peaks due to real part of the selfenergy have been neglected here for clarity.

to lie inside the sample, so that  $0 \leq y_0 = k_x c / eB \leq L$ . Hence,  $n_x^{max} = L^2 eB / (2\pi c)$  is the degeneracy of the Landau levels at a field  $B$ . Assuming  $L = 1$  mm and  $B = 10$  T the degeneracy is  $2.4 \times 10^9$ . For the Dirac limit,  $E_N = v_F \sqrt{2eBN/c}$ , assuming  $B = 10$  T and  $v_F = 6 \times 10^7$  cm/s,<sup>34</sup> we have  $E_N \approx 70\sqrt{N}$  meV, which is approximately  $1000\sqrt{N}$  K. This corresponds to a rather large spacing between Landau levels. On the other hand, for the Schrödinger limit we have  $E_N = \omega(N + 1/2)$  and for  $m = 0.1m_e$  and  $B = 10$  T this corresponds to a spacing of 11.6 meV or 150 K.

The surface states penetrate the bulk of the sample with their wave function falling off exponentially with the distance from the surface,  $\exp(-z/\lambda)$ . The penetration depth  $\lambda$  depends on the bulk properties of the system, i.e. the magnitude of the indirect hybridization gap,  $\Delta$ . In the Dirac limit, we can estimate  $\lambda$  through  $v_F/\lambda \approx \Delta$ , which for  $v_F = 6 \times 10^7$  cm/s and  $\Delta = 54$  K, yields  $\lambda \approx 85$  nm. In the Schrödinger limit, on the other hand,  $1/(2m\lambda^2) \approx \Delta$  and for  $m = 0.1m_e$  we have  $\lambda \approx 3$  nm.

Due to the finite size of the sample edge states are expected to appear. We have not considered the edge states here because they are not relevant to the NMR relaxation.

### III. RESULTS

#### A. NMR Hamiltonian

A local probe, such as NMR, is a useful tool to study the “in-gap” states of  $\text{SmB}_6$ . The  $^{11}\text{B}$  nuclei carry a nu-

clear total angular momentum  $I = 3/2$ . For a magnetic field parallel to a crystallographic axis there are two inequivalent sites in the  $B_6$  octahedra, which are denoted with B1 and B2 in Ref. 15. The quadrupolar splitting at the two sites yields a superposition of the spectra, so that to separate the lines it is convenient to measure the  $|3/2\rangle \leftrightarrow |1/2\rangle$  NMR transition.

We define spin-1/2 operators for the space spanned by the nuclear states  $|3/2\rangle$  and  $|1/2\rangle$ ,

$$\begin{aligned} S^+ &= |3/2\rangle\langle 1/2| \quad , \quad S^- = |1/2\rangle\langle 3/2| \quad , \\ S^z &= \frac{1}{2}(|3/2\rangle\langle 3/2| - |1/2\rangle\langle 1/2|) \quad . \end{aligned} \quad (6)$$

These operators satisfy the standard spin commutation relations,  $[S^\pm, S^z] = \mp S^\pm$  and  $[S^+, S^-] = 2S^z$ . The interaction of the nuclear spin with the topological surface states is via a contact exchange,  $\mathcal{H}_J = (J/2)[S^z\sigma_z + \frac{1}{2}(S^+\sigma_- + S^-\sigma_+)]\delta(\mathbf{r})$ .

SmB<sub>6</sub> is a cubic intermediate valent Kondo insulator and, according to specific heat data<sup>11</sup> and Raman scattering measurements,<sup>8</sup> the bulk ground wave function is a  $\Gamma_8$  quartet. At the surface the symmetry is tetragonal so that  $\Gamma_8$  is split into two Kramers doublets. The ground doublet can be parametrized by a pseudospin 1/2 and the two states are linear combinations of the true spin components. The Dirac Hamiltonian locks the true spin perpendicular to the momentum vector.<sup>21,33</sup> We assume that the exchange coupling of the nuclear spin is with the true spin. If the full  $f$ -states are considered, the orbital degree of freedom of the crystalline field Kramers doublet has to be traced out, yielding renormalizations of the expectation values of the Pauli matrices, which can be absorbed into the exchange coupling defined below.<sup>33</sup>

We now express the total Hamiltonian,  $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_Z + \mathcal{H}_J$ , where  $\mathcal{H}_Z$  is the Zeeman part, in terms of the eigenstates of  $\mathcal{H}_0$ . The two spinor components (true spin states) are denoted with a subindex 1 and 2. The creation and annihilation operators for states with energy  $E_N$  are  $c_{1,N-1,k}^\dagger$ ,  $c_{1,N-1,k}$  and  $c_{2,N,k}^\dagger$ ,  $c_{2,N,k}$ , respectively. Here  $N-1$  and  $N$  refers to the harmonic oscillator wave function and  $k$  is the degeneracy index  $k_x$  (we drop the subindex  $x$ ). The Hamiltonian has now the form

$$\begin{aligned} \mathcal{H}_0 &= \sum_{N,k} E_N \left[ c_{1,N-1,k}^\dagger c_{1,N-1,k} + c_{2,N,k}^\dagger c_{2,N,k} \right] , \\ \mathcal{H}_Z &= -(h_e/2) \sum_{N,k} \left[ c_{1,N-1,k}^\dagger c_{1,N-1,k} - c_{2,N,k}^\dagger c_{2,N,k} \right] , \\ \mathcal{H}_J &= \frac{J}{2} \sum_{N,k,N',k'} \left\{ S^z \left[ c_{1,N-1,k}^\dagger c_{1,N'-1,k'} \psi_{1,N-1,k}^* \right. \right. \\ &\quad \times \left. \psi_{1,N'-1,k'} - c_{2,N,k}^\dagger c_{2,N',k'} \psi_{2,N,k}^* \psi_{2,N',k'} \right] \\ &\quad + S^+ c_{2,N,k}^\dagger c_{1,N'-1,k'} \psi_{2,N,k}^* \psi_{1,N'-1,k'} \\ &\quad \left. + S^- c_{1,N-1,k}^\dagger c_{2,N',k'} \psi_{1,N-1,k}^* \psi_{2,N',k'} \right\} , \end{aligned} \quad (7)$$

where the normalized wave functions  $\psi$  are taken at  $x = y = 0$  (the resonating nucleus is at the origin) due to the

contact interaction, and  $h_e = g_{eff}\mu_B B$  with  $g_{eff}$  being the effective  $g$ -factor of the electrons.

## B. Knight shift

The Knight shift is the shift of the resonance due to the polarization of the conduction electrons. It renormalizes the external magnetic field,  $h = \gamma_N \mu_N B$ , to an effective field  $h' = h + \delta h'$ , where  $\gamma_N$  and  $\mu_N$  are the nuclear gyromagnetic factor and magneton, respectively. Here  $\delta h'$  is given by

$$\begin{aligned} \delta h' &= -J \sum_{N,k,N',k'} \left[ \langle c_{1,N-1,k}^\dagger c_{1,N'-1,k'} \rangle \psi_{1,N-1,k}^* \right. \\ &\quad \times \left. \psi_{1,N'-1,k'} - \langle c_{2,N,k}^\dagger c_{2,N',k'} \rangle \psi_{2,N,k}^* \psi_{2,N',k'} \right] \\ &= -\frac{JL}{2\pi} \sum_N \left[ f(E_N - h_e/2) \int dk |\psi_{1,N-1,k}|^2 \right. \\ &\quad \left. - f(E_N + h_e/2) \int dk |\psi_{2,N,k}|^2 \right] , \end{aligned} \quad (8)$$

where  $f(E)$  is the Fermi function. The harmonic oscillator wave functions are displaced by  $y_0(k)$  from the origin. We have then

$$\int dk |\psi_{2,N,k}|^2 = \frac{eB}{Lc} \int dy_0 |\psi_{2,N,k}(y_0)|^2 = \frac{eB}{Lc} , \quad (9)$$

and similarly for the other spinor component. To linear order in the magnetic field we have then

$$\delta h' \approx \frac{JeBh_e}{2\pi Tc} \sum_N f'(E_N) , \quad (10)$$

where  $f'$  is the derivative of the Fermi function. Hence, in the clean limit, where there are sharp Landau levels, the Knight shift is proportional to  $B$  and inversely proportional to  $T$ . The factor  $B$  arises from the degeneracy of the Landau levels,  $h_e$  from the Zeeman splitting of the surface states and  $T^{-1}$  is the Curie susceptibility of the Landau levels.

## C. Relaxation rate in a clean strong TKI

Assuming a Lorentzian, the dissipative part of the transversal dynamic spin susceptibility can be written as<sup>44</sup>

$$\chi_T''(\omega) = \frac{\omega/T_1}{(\omega - h')^2 + (1/T_1)^2} \chi_{T0} , \quad (11)$$

where  $\chi_{T0}$  is the static transversal susceptibility and  $T_1$  is the relaxation time. Perturbatively, the relaxation rate contributes to second order in the exchange  $J$ .

The transversal spin correlation function is given by  $\langle\langle S^+; S^- \rangle\rangle_\omega$ . It is related to the susceptibility by  $\chi_T(\omega) = -(\gamma_N \mu_N)^2 \langle\langle S^+; S^- \rangle\rangle_\omega$ . Equations of motion

for the imaginary part of this correlation function lead to

$$(\omega - h')^2 \langle \langle S^+; S^- \rangle \rangle''_\omega = \langle \langle j_c^+; j_c^- \rangle \rangle''_\omega, \quad (12)$$

where  $j^\pm$  is the spin current, given by<sup>44</sup>

$$\begin{aligned} j^+ &= [S^+, \mathcal{H}_J] = J \sum_{N,k,N',k'} \\ &\left\{ S^z c_{1,N-1,k}^\dagger c_{2,N',k'} \psi_{1,N-1,k}^* \psi_{2,N',k'} \right. \\ &- \frac{1}{2} S^+ \left[ c_{1,N-1,k}^\dagger c_{1,N'-1,k'} \psi_{1,N-1,k}^* \psi_{1,N'-1,k'} \right. \\ &\left. \left. - c_{2,N,k}^\dagger c_{2,N',k'} \psi_{2,N,k}^* \psi_{2,N',k'} \right] \right\}. \end{aligned} \quad (13)$$

The subindex  $c$  denotes the cumulant contraction (Hartree-Fock factorization) that gives rise to the Knight shift  $\delta h'$  discussed in section III.B (Eq. (8)). Inserting the spin current operator, the correlation function to second order in  $J$  yields

$$\begin{aligned} \langle \langle j_c^+; j_c^- \rangle \rangle''_\omega &= J^2 \sum_{N,k,N',k',\bar{N},\bar{k},\bar{N}',\bar{k}'} \\ &\left\{ \psi_{1,N-1,k}^* \psi_{2,N',k'} \psi_{2,\bar{N},\bar{k}}^* \psi_{1,\bar{N}',\bar{k}'} \right. \\ &\times \langle \langle S^z c_{1,N-1,k}^\dagger c_{2,N',k'}; S^z c_{2,\bar{N},\bar{k}}^\dagger c_{1,\bar{N}',\bar{k}'} \rangle \rangle''_\omega \\ &+ \frac{1}{4} \psi_{1,N-1,k}^* \psi_{1,N'-1,k'} \psi_{1,\bar{N}-1,\bar{k}}^* \psi_{1,\bar{N}'-1,\bar{k}'} \\ &\times \langle \langle S^+ c_{1,N-1,k}^\dagger c_{1,N'-1,k'}; S^- c_{1,\bar{N}-1,\bar{k}}^\dagger c_{1,\bar{N}'-1,\bar{k}'} \rangle \rangle''_\omega \\ &+ \frac{1}{4} \psi_{2,N,k}^* \psi_{2,N',k'} \psi_{2,\bar{N},\bar{k}}^* \psi_{2,\bar{N}',\bar{k}'} \\ &\times \langle \langle S^+ c_{2,N,k}^\dagger c_{2,N',k'}; S^- c_{2,\bar{N},\bar{k}}^\dagger c_{2,\bar{N}',\bar{k}'} \rangle \rangle''_\omega \left. \right\}. \end{aligned} \quad (14)$$

The correlation functions are now evaluated to zero-order in  $J$  using  $\mathcal{H}_0 + \mathcal{H}_Z$

$$\begin{aligned} \langle \langle j_c^+; j_c^- \rangle \rangle''_\omega &= -\frac{\pi J^2}{4} \sum_{N,N',k,k'} \left\{ \delta(E_N - E_{N'} + \omega - h_e) \right. \\ &\times |\psi_{1,N-1,k}|^2 |\psi_{2,N',k'}|^2 (n_{3/2} + n_{1/2}) \\ &\times [f(E_N - h_e/2) - f(E_{N'} + h_e/2)] \\ &+ 2\delta(E_N - E_{N'} + \omega - h') \left[ |\psi_{1,N-1,k}|^2 |\psi_{1,N'-1,k'}|^2 \right. \\ &\times [n_{3/2} f(E_N - h_e/2)(1 - f(E_{N'} - h_e/2)) - n_{1/2} \\ &\times f(E_{N'} - h_e/2)(1 - f(E_N - h_e/2))] + |\psi_{2,N,k}|^2 \\ &\times |\psi_{2,N',k'}|^2 [n_{3/2} f(E_N + h_e/2)(1 - f(E_{N'} + h_e/2)) \\ &\left. \left. - n_{1/2} f(E_{N'} + h_e/2)(1 - f(E_N + h_e/2)) \right] \right\}, \end{aligned} \quad (15)$$

where  $n_{3/2}$  and  $n_{1/2}$  are the occupation numbers of the nuclear spin states. The sums over  $k$  and  $k'$  can be transformed into integrals and using the orthonormality of the wave functions, Eq. (9), we obtain

$$\langle \langle j_c^+; j_c^- \rangle \rangle''_\omega = -\frac{\pi J^2}{4} \left( \frac{eB}{2\pi c} \right)^2 \sum_{N,N'} \left\{ (n_{3/2} + n_{1/2}) \right.$$

$$\begin{aligned} &\times \delta(E_N - E_{N'} + \omega - h_e) [f(E_N - h_e/2) \\ &- f(E_{N'} + h_e/2)] + \delta(E_N - E_{N'} + \omega - h') \sum_{\sigma=\pm 1} \left[ n_{3/2} \right. \\ &\times f(E_N + \sigma h_e/2)(1 - f(E_{N'} + \sigma h_e/2)) \\ &\left. - n_{1/2} f(E_{N'} + \sigma h_e/2)(1 - f(E_N + \sigma h_e/2)) \right] \left. \right\}. \end{aligned} \quad (16)$$

Through Eqs. (11) and (12) we have that to second order in  $J$  ( $\omega$  is to be taken at the resonance frequency  $h'$ )

$$\frac{1}{T_1} = -(\gamma_N \mu_N)^2 \langle \langle j_c^+; j_c^- \rangle \rangle''_{\omega=h'} / (h' \chi_{T0}). \quad (17)$$

Due to the delta functions, the expression (16) vanishes unless  $E_N - E_{N'} + \omega - h_e = 0$  or  $E_N - E_{N'} + \omega - h' = 0$  for some  $N$  and  $N'$ .

In a normal metal the NMR relaxation rate is given by the Korringa relaxation, i.e. it is proportional to  $T$  and the square of the exchange coupling to the  $s$ -wave conduction states (at the site of the NMR ion) times their density of states. It follows that Eq. (17) does not lead to a Korringa-like relaxation unless the conduction states have a continuum energy spectrum. Hence, there is no Korringa-like NMR relaxation with clean surface states. Similarly, the electrical resistivity is only going to be nonzero if there is a continuum energy spectrum. This is not the case for a clean surface in a magnetic field, where the energy states are sharp and discrete.

The delta functions in Eq. (16) are indicative of a formation of a bound state of the nuclear spin with the Landau level closest to the Fermi level. The spin degree of freedom then resonates within this bound state without being able to relax into a continuum.

#### D. Strong TKI with dirty surfaces

The surfaces of a realistic sample have a considerable amount of defects. Defects can be magnetic impurities or nonmagnetic scatterers. Magnetic impurities break the time reversal symmetry and lift the topological protection of the surface states. We consider here nonmagnetic defects, such as adsorbed nonmagnetic atoms, missing Sm or B atoms, surface steps, etc. that do not break the time reversal symmetry, but only the translational invariance. As a consequence of the scattering off the defects, the surface quasi-particles acquire a finite linewidth and the Landau levels are no longer perfectly sharp. The density of states has then an energy continuum.

The broadening of electron Landau levels in 2D due to light disorder has been studied previously in the context of the 2D electron gas<sup>45-47</sup> using a selfconsistent averaging over the positions of the defects. The method is now known as the fully self-consistent Born approximation (FSBA) and has been extended to study the effect of impurity scattering on the magnetoresistance of graphene.<sup>43,48</sup> This extension involves a Dirac cone dispersion, similar to the present problem. The real part

of the self-consistent approximation for the electron self-energy yields a shift of the Landau level energy and the imaginary part a linewidth of the state. The real part is not of primary relevance for the present purposes and the imaginary part is approximately constant over the energy range  $h_e$  ( $\approx 1$  meV) under consideration. The density of states for a Landau level, which is a delta function in the absence of defects, is now constant over the relevant energy interval.

For each Landau level we introduce  $\int d\epsilon \delta(\epsilon - E_N) = 1$  and replace the  $\delta$ -function by a broadened density of states  $\int d\epsilon \rho(\epsilon - E_N)$  in Eq. (8) for the Knight shift. Assuming that  $\rho$  is constant over a small interval about  $E_N$  (of the order of  $h_e$ ) we can carry out the  $\epsilon$  integration. Denoting with  $\rho_N$  the density of states at the Landau level  $N$ , we arrive at

$$\delta h' \approx -\frac{JeBh_e}{2\pi c} \sum_N \rho_N. \quad (18)$$

Hence, in the dirty limit the Knight shift is still proportional to  $B$ , arising from the degeneracy of the Landau levels, but no longer proportional to  $1/T$ . The most relevant term in Eq. (18) is for  $N^*$ , where  $N^*$  is the Landau level pinning the Fermi level.

For the relaxation rate we again insert  $\int d\epsilon \rho(\epsilon - E_N)$  for each Landau level. The Zeeman splitting of the conduction states does not play a relevant role for this case so that to simplify we consider  $h_e = 0$ . From Eq. (16) we have then for the leading contribution to the spin-current correlation function

$$\begin{aligned} \langle\langle j_c^+; j_c^- \rangle\rangle''_{\omega} &= -\frac{\pi J^2}{4} \left(\frac{eB}{2\pi c}\right)^2 \sum_{N, N'} \int d\epsilon \rho(\epsilon - E_N) \times \\ &\int d\epsilon' \rho(\epsilon' - E_{N'}) \left\{ \delta(\omega - \epsilon' + \epsilon)(n_{3/2} + n_{1/2}) \times \right. \\ &[f(\epsilon) - f(\epsilon')] + 2\delta(\omega - h' - \epsilon' + \epsilon) \left[ n_{3/2} f(\epsilon)(1 - f(\epsilon')) \right. \\ &\left. \left. - n_{1/2} f(\epsilon')(1 - f(\epsilon)) \right] \right\}. \end{aligned} \quad (19)$$

After integrating over  $\epsilon'$  we obtain

$$\begin{aligned} \langle\langle j_c^+; j_c^- \rangle\rangle''_{\omega} &= -\frac{\pi J^2}{4} \left(\frac{eB}{2\pi c}\right)^2 \sum_{N, N'} \int d\epsilon \rho(\epsilon - E_N) \times \\ &\left\{ \rho(\epsilon - E_{N'} + \omega)(n_{3/2} + n_{1/2})[f(\epsilon) - f(\epsilon + \omega)] \right. \\ &+ 2\rho(\epsilon - E_{N'} + \omega - h') \left[ n_{3/2} f(\epsilon)(1 - f(\epsilon + \omega - h')) \right. \\ &\left. \left. - n_{1/2} f(\epsilon + \omega - h')(1 - f(\epsilon)) \right] \right\}. \end{aligned} \quad (20)$$

Assuming that  $\rho$  is constant over a small interval about  $E_N$  we can carry out the  $\epsilon$  integration. Denoting again with  $\rho_N$  the density of states at the Landau level  $N$ , we arrive at

$$\langle\langle j_c^+; j_c^- \rangle\rangle''_{\omega} = -\frac{\pi J^2}{4} \left(\frac{eB}{2\pi c}\right)^2 \sum_{N, N'} \rho_N \rho_{N'} \left\{ \right.$$

$$\begin{aligned} &\omega(n_{3/2} + n_{1/2}) + 2n_{3/2}(\omega - h') \frac{\exp((\omega - h')/T)}{\exp((\omega - h')/T) - 1} \\ &\left. - 2n_{1/2}(\omega - h') \frac{1}{\exp((\omega - h')/T) - 1} \right\}. \end{aligned} \quad (21)$$

The static transversal susceptibility for the free nuclear spin is  $\chi_{T0} = (\gamma_N \mu_N)^2 (n_{3/2} + n_{1/2})/h'$ , so that

$$\begin{aligned} \frac{1}{T_1} &= -(\gamma_N \mu_N)^2 \langle\langle j_c^+; j_c^- \rangle\rangle''_{\omega=h'}/(h' \chi_{T0}) \\ &= \frac{\pi}{4} \left(\frac{eBJ}{2\pi c}\right)^2 \sum_{N, N'} \rho_N \rho_{N'} \left[ 2T + h' \frac{n_{3/2} + n_{1/2}}{n_{3/2} - n_{1/2}} \right] \\ &\approx \pi T \left(\frac{eBJ}{2\pi c}\right)^2 \sum_{N, N'} \rho_N \rho_{N'}, \end{aligned} \quad (22)$$

where we used that  $(n_{3/2} + n_{1/2})/(n_{3/2} - n_{1/2}) = \coth(h'/2T) \approx 2T/h'$ , since  $h' \ll T$ . Hence, the relaxation rate for nuclei at the surface of a mildly dirty crystal is Korringa-like. The relaxation rate grows linearly with temperature and quadratically with the magnetic field. There is an additional magnetic field dependence arising from the density of states of the Landau levels. The dominant term in Eq. (22) is for  $N = N' = N^*$ , where  $N^*$  is the Landau level pinning the Fermi level,

$$\frac{1}{T_1} \approx \pi T \left(\frac{eBJ}{2\pi c}\right)^2 \rho_{N^*}. \quad (23)$$

We are not presenting an explicit calculation of the self-energy due to defect scattering using the FSBA. The procedure would be analogous to the one described in Ref. 48 for the case of graphene. It involves several unknown parameters, such as the density of impurities (assumed to randomly distributed) and the scattering strength by the defects. In contrast the parameters of the clean surface can be estimated from experiments and first principle calculations. The density of states is obtained numerically by solving coupled equations. Since the dependence of the surface states on roughness<sup>32,37</sup> is still an open experimental issue and the input parameters for the calculation are unknown, an explicit calculation is not going to add much to the understanding of the problem.

In Fig. 1(b) and 1(c) we show the density of states with weak impurity scattering and in the dirty limit, respectively. Here the imaginary part of the selfenergy was assumed to be constant (Lorentzians) and the real part of the selfenergy, responsible for the shifts of the positions of the peaks, has been neglected.

#### IV. CONCLUSIONS

We have studied the NMR Korringa relaxation and Knight shift of  $^{11}\text{B}$  nuclei in the topological Kondo insulator  $\text{SmB}_6$ . Experimental and theoretical evidence indicates that the bulk is gapped at low  $T$  and only the

surface states contribute to electrical conduction. The observed Korringa relaxation is then expected to arise from the surface conduction states. In a magnetic field the clean 2D electron gas has a discrete energy spectrum as a consequence of the Landau quantization. We have shown that the Knight shift is proportional to  $B/T$  and the relaxation rate is not Korringa-like in this scenario, which contradicts the experimental evidence.<sup>15</sup>

If the surface has nonmagnetic defects, the scattering of the conduction states off the defects gives rise to a line width of the Landau levels. The density of states then consists of peaks with finite width rather than delta functions as for the clean surface. A small broadening (of the order of 1 meV) is sufficient to yield a Korringa rate proportional to the temperature. The proportionality constant is field dependent, since the degeneracy of the Landau levels is proportional to the field.

Since only electronic surface states participate in the relaxation process of the  $^{11}\text{B}$  nuclei, this relaxation is then expected to be weak. Since the wave function falls off exponentially into the crystal, depending on their distance from the surface the  $^{11}\text{B}$  nuclei should have different  $T_1$  with  $T_1$  becoming really large in the insulating part of the crystal. Hence, one should expect then a superposition of several relaxation times arising from the different layers. This gives rise to a “stretched” exponential time dependence. Although there is some evidence for a small stretching, the experimental data is consistent with a single relaxation time.<sup>15</sup>

For the sake of simplicity in the present calculation we considered only one Dirac core. Band structure calculations<sup>24</sup> (see also Ref. 49 for the electronically similar compound  $\text{PuB}_6$ ) suggest that there are three Dirac cones, one at the  $\bar{\Gamma}$ -point and the other two at the  $\bar{X}(\bar{Y})$ -points of the surface Brillouin zone. The latter two are symmetry-equivalent. The energies of the cone-vertices are not protected and could shift with the sample preparation, e.g. with the disorder on the surface. The exchange Hamiltonian of the of the nuclear spin with the conduction states,  $\mathcal{H}_J$ , consists then of three terms of the type described in Eq. (7) with different hyperfine exchange couplings. The Knight shift and the relaxation rate are then the superposition of the contributions of the three bands. We conclude that there is no qualitative change of the results if several bands are considered in comparison to the single band results.

Furthermore, we have assumed that the surface electrons do not interact with each other. Since the kinetic energy of the Dirac particles is relatively small, interactions could play a relevant role at very low  $T$ .<sup>50</sup> Experimentally ferromagnetic long-range order has been found at temperatures far below the  $T$ -range of the NMR measurements.<sup>51</sup> Edge states originating from the finite size of the sample have also been neglected.

In a standard NMR experiment in a metallic system the Korringa product, defined as  $\kappa = [T_1 T (\delta h'/B)^2]^{-1}$  where  $\delta h'/B$  is the Knight-shift constant, is a constant. This is not the case in  $\text{SmB}_6$ , where below 10 K  $\kappa$  is

strongly enhanced and strongly magnetic field and temperature dependent (see Fig. 4 in Ref. [15]).

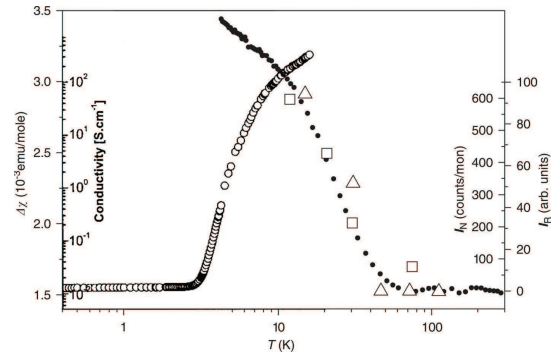


FIG. 2: The contribution of the “in-gap” state to the susceptibility ( $\Delta\chi$ ) (solid circles),<sup>12</sup> the temperature dependence of the 16 meV magnetic excitation intensities measured by inelastic neutron scattering  $I_N$  (open squares),<sup>9</sup> and the intensity of the Raman transition  $I_R$  (open triangles)<sup>8</sup> compared to electrical conductivity data (open small circles) from Ref. 52. While the surface states contributing to the conductivity are active below 4 K, the “in-gap” states are formed at much higher  $T$ . (Figure adapted from Ref. 9).

While the low  $T$  resistivity of  $\text{SmB}_6$  saturates below 4 K, which is attributed to the topological surface states, the onset of the in-gap states in the susceptibility<sup>12</sup>, in inelastic neutron scattering<sup>9</sup>, the NMR Knight shift and relaxation<sup>15</sup> and Raman spectroscopy<sup>8</sup> occurs at higher  $T$ , of the order of 25 - 30 K. This is shown in Fig. 2. This suggests the possibility of an alternate mechanism to the surface states for the magnetic properties. The intensity of the observed effects also appears to be too large to be attributed solely to surface states. A model to explain the in-gap bound states in Kondo insulators has been proposed by Kasuya using a localized Kondo model.<sup>53</sup> The lowest excitation state is an  $s$ -wave exciton, leading to predictions consistent with neutron scattering results. Along similar lines Riseborough<sup>16</sup> proposed that the mixed valence nature of the system gives rise to antiferromagnetic correlations leading to in-gap magnetic excitations, analogous to antiparamagnons in a metal.<sup>54</sup> These magnetic exciton-like states exist in the bulk but do not contribute to the electrical conductivity, since an electron and a hole are bound in a pair and the bound state has charge zero. Hence, the electrical conductivity is a property of the surface states. The in-gap bound states, however, have their own temperature and field dependence, which should reflect in the magnetic susceptibility and NMR properties. In particular, at low  $T$  the relaxation rate displays a broad peak that moves to higher temperatures with increasing field, that could be the signature of such a bound state (see Fig. 3). The intensity of the peak dramatically decreases with magnetic field, possibly due to the quenching of the antiferromagnetic correlations. A very recent calculation of the in-gap collective mode spectrum of  $\text{SmB}_6$  can be found in Ref.



55.

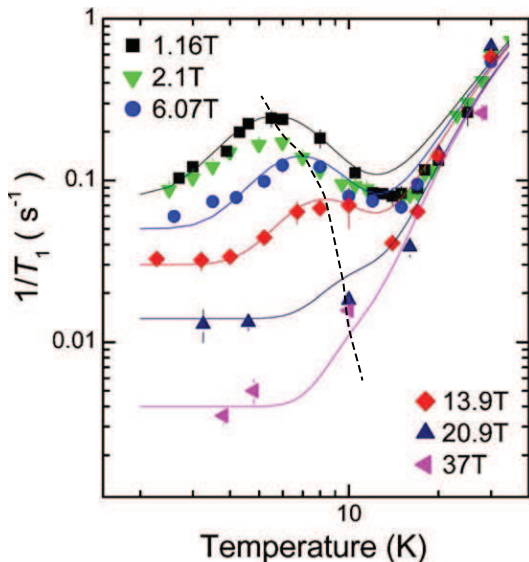


FIG. 3: (Color online) Low temperature spin lattice relaxation rates as a function of  $T$  for different magnetic fields (adapted from Ref. 15). The solid lines are fits to a simple model for “in-gap” states<sup>15</sup> and can be considered as guides to the eye. The data shows peaks or shoulders as a function of  $T$  and dashed curve interpolates between these maxima.

It would be of interest to carry out the NMR measurements on samples of different thickness. A measured relaxation rate that is independent of the sample thickness would be convincing evidence for a Korringa relaxation into surface states and exclude a magnetic exciton mechanism. Low temperature ( $T = 1.6$  K and 3.8 K) X-band Electron Spin Resonance (ESR) of Sm in SmB<sub>6</sub> shows evidence for several transitions,<sup>56</sup> which have been interpreted as arising from the  $\Gamma_8$  quadruplet of Sm<sup>3+</sup> defects and  $\Gamma_6$ -states in the crystal. Earlier measurements of Gd impurities in SmB<sub>6</sub> have been presented in Ref. 57. Due to the intermediate valence of SmB<sub>6</sub> the Sm ions in the

bulk are not expected to resonate. In the light of TKIs it would be desirable to repeat these measurements on cleaner samples and higher frequencies than X-band to see if the cyclotron resonances of the electron gas can be observed.

In summary, there are numerous problems with the interpretation of the NMR data. (1) Magnetic data suggest the existence of two energy scales, one associated with the topological surface states and the other with the formation of bulk in-gap states, possibly exciton-like bound states. (2) If the NMR relaxation is due to the topological surface states the contribution of the different surface layers would give a different  $T_1$  depending on the distance from the surface. This would give rise to a strongly “stretched” exponential time dependence. Although there is evidence for some stretching the experiments are consistent with a single relaxation time. In addition the intensity of the resonance line would be faint since only <sup>11</sup>B nuclei at the surface would be able to resonate. (3) Under optimal conditions the sensitivity of an NMR experiment requires the detection of  $10^{15}$  spins. The number of surface <sup>11</sup>B atoms in a  $2 \times 2 \times 1$  mm<sup>3</sup> single crystal is roughly  $10^{15}$ , i.e. barely within the limits of detection. (4) At low  $T$  the relaxation rate displays a broad peak that moves to higher temperatures with increasing field as shown in Fig. 3 by the dashed line. This peak could be the signature of the excitonic bound state in the relaxation rate. Although none of the above findings is conclusive, they are evidence suggesting different mechanisms for the electric transport and the magnetic properties.

### Acknowledgments

The author would like to thank Dr. Arneil Reyes for helpful discussions. The support by the U.S. Department of Energy under grant DE-FG02-98ER45707 is acknowledged.

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