Spin-exciton and topology in SmB$_6$

W. T. Fuhrman$^{1,1}$, J. Leiner$^{2,2}$, P. Nikolić$^{1,3}$, G. E. Granroth$^4$, M. B. Stone$^2$, M. D. Lumsden$^2$, L. DeBeer-Schmitt$^5$, P. A. Alekseev$^{6,7}$, J.-M. Mignot$^8$, S. M. Koohpayeh$^1$, P. Cottingham$^{1,9}$, W. Adam Phelan$^{1,9}$, L. Schoop$^{1,9}$, T. M. McQueen$^{1,10}$, and C. Broholm$^{2,1}$

$^1$Institute for Quantum Matter and Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218 USA
$^2$Quantum Condensed Matter Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
$^3$School of Physics, Astronomy and Computational Sciences, George Mason University, Fairfax, VA 22030, USA
$^4$Neutron Data Analysis and Visualization Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
$^5$Instrument Source Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
$^6$National Research Centre “Kurchatov Institute”, 123182 Moscow, Russia
$^7$National Research Nuclear University "MEPhI", 115409 Moscow, Russia
$^8$Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, France
$^9$Department of Chemistry, The Johns Hopkins University, Baltimore, Maryland 21218 USA
$^{10}$Department of Chemistry, Princeton University, Princeton, NJ 08540

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Using inelastic neutron scattering, we map a 14 meV resonant mode in SmB$_6$ and describe its relation to the low energy insulating band structure. The resonant intensity is sharply confined to the X and R high symmetry points and disperses less than 2 meV through the zone. Repeating outside the first Brillouin zone, the mode is coherent with a 5$d$-like magnetic form factor. We describe how band inversion in cubic symmetry can be inferred from neutron scattering and show that a slave boson treatment of a 3rd-neighbor dominated hybridized 2 species ($d/f$) band structure can produce a spin exciton with the observed characteristics.

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Recent theoretical work suggests SmB$_6$ could be a Topological Kondo Insulator (TKI), with an insulating bulk at low temperatures and topologically protected metallic surface states [1-7], previously ascribed to impurities [8]. Because electron-electron interactions rather than a free fermion band structure are responsible for this insulator, SmB$_6$ has garnered great interest. Experiments generally support the theoretical proposal [9-15], however, information about the band structure is limited due to the polar surface and the low energy scale. In this letter we show that a full momentum space map of inelastic magnetic neutron scattering provides valuable information about pseudo-nesting conditions for the renormalized band structure and supports a topologically non-trivial band structure for SmB$_6$.

The low energy magnetic neutron scattering cross section for SmB$_6$ is dominated by a resonant mode near 14 meV with bandwidth < 2 meV. Previous work discovered intensity at a single momenta, R $[(\frac{1}{2} \frac{1}{2} \frac{1}{2})]$, and thoroughly investigated it with respect to temperature dependence, doping, and anisotropy [16,21]. In this work we show the mode is also intense near the X $[(\frac{3}{2} \frac{1}{2} 0)]$ point and present, albeit dramatically weaker, beyond the first zone. We use this observation to develop a minimal band structure based on a dominant third-neighbor interaction in a hybridized tight-binding model similar to early two-band theoretical treatments [22-25], and infer general connections between the wave vector dependence of magnetic neutron scattering and band inversion. We model both X and R point intensity in a slave-boson approach, resulting in the the formation of an interaction-protected bound state with dispersion similar to the experiment.

SmB$_6$ has Pm3n symmetry with an octahedron of Boron in the center of the simple cubic unit cell. A single crystal of SmB$_6$ grown by the floating zone method using the non-neutron-absorbing isotopes $^{154}$Sm and $^{111}$B (mozaicity of 25') was synthesized by Yu Paderno and E. Konovalova and initially adopted for lattice [26] and magnetic [21] dynamics studies on triple-axis spectrometers. This experiment was carried out on the SEQUOIA time of flight spectrometer at the SNS using incident energies of 50 meV, 80 meV, and 100 meV with energy resolution better than 2 meV; the 14 meV range was measured for momentum transfer down to 0.5 Å$^{-1}$ and energy resolution of 1.8 meV [27,29]. Intensity was scaled to absolute units for the differential scattering cross section by normalizing to neutron scattering from acoustic phonons and Bragg peaks [30].

Fig. 1 shows the $Q$-dependence of the inelastic scattering intensity, integrated from 12 meV to 16 meV. Visible at the R point is the intensity maximum previously interpreted as an excitation from the spectrum of an intermediate-radius exciton [23], formed due to the mixed valence state of Sm. The excellent small scattering angle capabilities of SEQUOIA now brings a strong peak at the X point into view, which is replicated be-
FIG. 1. Energy integrated intensity (a, b, d) of high symmetry planes. (a) $^{154}$Sm$^{11}$B$_6$ at 5K. (b) La$^{11}$B$_6$ at 5K. (c) Crystal structure and smallest unique portion of the Brillouin zone (d) $Q$-dependence of a Lindhard-type function for $\chi_0$. (e) Band structure of the $(h k 0)$ plane. Translation from X to M shows the change in band character. Inset, schematic representation of pseudo-nesting vectors from hybridized bands with opposite dispersions. (f) Fundamental processes in the perturbative slave-boson theory. The slave-boson-mediated conversion between an f and a d electron(1) dominates over $f$ electron scattering on slave bosons (2). This resonant conversion provides electron-hole pairing glue, stabilizing a bound state (3). Self-energy corrections (4) shrink the exciton bandwidth and produce the relatively flat collective mode seen in the experiment. The self-consistently renormalized slave-boson propagator in (4) stands for all wavy lines in diagrams of (3); its numerical properties are extracted from experimental data by a fitting procedure.

FIG. 2. Neutron scattering cross section for SmB$_6$ at 5 K along high symmetry directions (inset). Dashed line shows the dispersion calculated via slave boson renormalization.

yond of the first zone at $X+G = (\frac{1}{2}\text{10})$. The intensity is greatly diminished beyond the first Brillouin zone, indicating the associated spin-density extends beyond the 4f orbital (Fig. 4).

Fig. 2 shows $Q$-dependent spectrum of neutron scattering intensity along high-symmetry paths though the Brillouin zone. Intensity is confined to regions near the X and R points where the mode energy is minimal. The overall bandwidth of the resonance is less than 2 meV.

Fig. 3 provides a quantitative overview of the resonant mode. All peaks versus energy are resolution-limited (dashed line in (c)), indicating a long lived collective mode that is isolated from the continuous electron hole pair continuum. The oscillator strength half-way between X and R falls to less than 20% of peak values without significant broadening (Fig. 3(b)). This confinement in momentum space contrasts with a conventional crystal field exciton for which the oscillator strength is $Q$-independent.

Fig. 4 shows the energy integrated intensity of the resonance at X and R type reciprocal lattice points within and beyond the first Brillouin zone. For reference, Fig. 4(a) shows the wave vector dependence of nuclear Bragg intensities normalized to the corresponding squared structure factor following identical symmetrization and integration procedures as for the magnetic scattering. The $Q$-independence of these data shows the experiment and analysis procedures accurately measure the $Q$-dependence of the cross section despite the effects
of absorption and crystal mosaic.

When the magnetic ion forms a simple Bravais lattice, as for SmB$_6$, Bloch’s theorem forms the differential wave function $I(q + G) = I(q) |F(q + G)|^2$, where $F(Q) = <J_0 > + (1 - \frac{1}{2})<J_2>$, and $<J_n> = \int d^3 r J_n(\mathbf{r})^2 J_n(\mathbf{q} \cdot \mathbf{r})$. $J_n$ is the $n$th spherical Bessel function and $g$ is the Landé factor. We compare the experimental result to the form-factors of potential magnetic scattering centers. Samarium is of mixed valence, with magnetism resulting from the Sm$^{3+}$ (J= $\frac{5}{2}$) state; however, the data is inconsistent with the intermediate-valence (IV) form factor from polarized neutron studies. The B$_6$ octahedron would be a magnetic scattering center if the origin of the scattering were electron transfer (Sm$^{2+}$ (B$_6$)$^{2-}$ and Sm$^{3+}$ (B$_6$)$^{3-}$); this can be ruled out as the corresponding (B$_6$)$^{3-}$ form factor is indistinguishable from zero beyond the first Brillouin zone, while we observe resonance intensity at X+G. Instead, the data follows the 5$d$ electron form factor, indicating a critical role for such orbitals in the exciton.

Integrating the exciton scattering over a full Brillouin Zone elucidates the relative contribution of the exciton to the total moment via the sum rule: $\frac{1}{2}g^2 J(J + 1) = \int \int S^<^2(Q,\omega)d\omega dQ$. Summing the intensity from 12 meV to 16 meV recovers $\approx 29(3)$% of the total effective moment squared of $(2.52 \mu_B)^2$ so the exciton described by a 5$d$-type form factor carries a significant portion of the samarium dipole moment.

Because the wave vector dependence of the magnetic neutron scattering detected suggests interpretation in terms of a band picture, we proceed to develop a minimal phenomenological model. The nearest electron density to samarium is the B$_6$ cluster. The molecular orbital diagram of the non-magnetic B$_6^{2-}$ cluster has its lowest unoccupied molecular orbitals on opposing faces of the octahedra in a t$_{1_u}$ state, allowing for efficient super-exchange along the body diagonal in the magnetic Sm$^{3+}$ (B$_6$)$^{3-}$ state, and so we consider a band structure with only 3rd neighbor hopping.

Although the chemical potential lies in a gap so there is no Fermi surface and no nesting in the conventional sense, 5$d$-electron “pseudo nesting” (PN) can still enhance finite energy magnetic excitations through inter-band transitions. X and R PN is inherent to a wide range of tight binding band structures dominated by 3rd neighbor hopping.

The 4$f$-bands may likewise be assumed dominated by third neighbor hopping albeit with a much smaller bandwidth. To retain a full insulating gap under $f$ – $d$ hybridization, the hopping amplitude for $f$ electrons must have the opposite sign from $d$ electrons. This ensures the bands move away from the chemical potential beyond their intersection points, forming a gap while extrema form in the hybridized bands near intersections (inset to Fig. 4(c)). The corresponding inter-band transitions now yield PN. An X-type PN boundary is for example visible in Fig. 1 between regions of hybridization.
The corresponding phenomenological band structure contains deep band-inversion pockets around the X-points and a gap of 15 meV, consistent with ARPES [12]. Due to the 4-fold degeneracy of the bands at the Γ and R points, only the X and M points can contribute to the 3D topological invariant [30], so the proposed phenomenological band structure is topologically non-trivial. The TKI nature is in fact a direct consequence of the opposite signs of the dominating third near neighbor hopping amplitudes for 4f and 5d electrons.

When modulated by the 5d electron form factor, the static susceptibility calculated from the resultant particle-hole Green’s function is consistent with the wave vector dependent intensity of the resonant mode, Fig. 1(d). No scale factor between X and R was required, an indication of similar density of states for both types of PN wave vectors. In our 3rd neighbor model, both X and R intensity result from PN between cubic faces and as such have nearly identical DOS. Thus the experimental results in Fig. 1(a) and 1(b) provide detailed support for dominant 3rd neighbor hopping.

Informed by this analysis we recognize that under high symmetry conditions the wave vector dependence of inelastic magnetic neutron scattering provides information about band topology. The 14 meV spin-exciton we have observed is associated with transitions across the hybridization gap where sharply dispersing d-bands define inversion pocket boundaries (Fig. 1(e)). The symmetry of any such transition must be matched by its corresponding pseudo Fermi surface, a condition manifestly satisfied for pockets centered on a HSP with the same symmetry as the observed transfer. Any deviation from such placement would require the pocket have the additional symmetries of its host HSP. Thus, an observation of strong intensity at X without intensity along M (i.e. the (110) direction) puts strong constraints on the location of the pocket outside of the X point, while the absence of intensity along M similarly precludes a pocket at M. Since in cubic Kondo TI only the X and M points can contribute to the topological invariant [30], the observation of magnetic intensity at X but not at M indicates a topologically non-trivial hybridized band structure for SmB$_6$. Comprehensive neutron scattering data combined with such reasoning may facilitate analysis of other potential TKI such as YbB$_6$ and PuB$_6$ [33, 36].

The collective mode we observed can be understood as an exciton created by Coulomb interactions and protected against decay by the hybridization bandgap. Exciton formation is efficient because the bandgap is narrow and the bandwidth of f orbitals is much smaller than the interaction energy scale. The minimal second-quantized Hamiltonian of SmB$_6$ is formulated on the lattice of Sm atoms:

$$H = \int_{IBZ} \frac{d^3k}{(2\pi)^3} \left[ \sum_{\sigma} \epsilon_k d^\dagger_{k\sigma} d_{k\sigma} + \sum_{\alpha} \epsilon_{\alpha k} f^\dagger_{\alpha k} f_{\alpha k} \right] + \sum_{\alpha\beta R} \left[ V_{\alpha k} d^\dagger_{\alpha k} f_{\alpha k'} + h.c. \right] + U \sum_{\alpha\beta R} f^\dagger_{\alpha R} f_{\alpha R} f^\dagger_{\beta R} f_{\beta R} ,$$

where $d_{\alpha k}$ are d-electron field operators indexed by spin $\sigma \in \{\uparrow, \downarrow\}$, and $f_{\alpha k}$ are f-electron operators labeled by the crystal-field multiplet index $\alpha$, which takes into account strong spin-orbit coupling within Sm. Crystal fields introduce hybridization $V$ which produces the narrow bandgap. Coulomb interaction is most influential on the narrow bandwidth f electrons, suppressing double occupancy. We thus model interactions by on-site repulsion $U$ among f electrons only. The slave-boson approximation ($U \to \infty$), removes the interaction term in favor of an explicit no-double-occupancy constraint imposed on every site with the help of an auxiliary slave boson field. The quantum fluctuations of slave bosons renormalize the spectrum and give rise to exciton pairing. These effects can be calculated perturbatively using the random-phase approximation [37].

The perturbation theory is built on top of a mean-field condensate of slave bosons, which formally plays the main role in renormalizing and shrinking the hybridization band gap. Slave boson fluctuations introduce further renormalizations of the bandstructure, which we neglect, and provide the pairing glue for the excitons, Fig. 1(f), which we retain. Fig. 1(f) shows the associated Feynman diagrams which will be discussed in detail elsewhere [38].

Fig. 2 compares our experimental results with the calculated exciton dispersion. Since the precise microscopic values of parameters are not known, we fit their renormalized values to phenomenologically match the calculated and measured spectra. Using the band structure described above, the calculated exciton dispersion relation is consistent with the experiment, having comparable bandwidth and minima at HSP. The existence of the exciton and its apparent origin in Coulomb interactions portray SmB$_6$ as a correlated (Mott) insulator where the lowest energy excitations are bosonic rather than fermionic as in band-insulators.

We observed a 14 meV collective mode in an extensive region of momentum-space and describe a minimal model where a spin-exciton bound state is formed by interactions under slave-boson renormalization. This long-lived mode is a consequence of the protection afforded by correlations within an insulator born of hybridization; the Kondo singlet fluctuations it represents is a testament to the strong correlations driving the TI phase in this compound and may lead the way to yet more exotic topological physics. The observed form-factor is evidence for a significant role of 5d orbitals in the exciton, and the symmetry of the high intensity regions in momentum space reflects an underlying topologically non-trivial
band structure in SmB$_6$.

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* W.T.F. and J.L. contributed equally to this work.

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