Supplementary Information for ‘Magnetic field dependence of the spin-1/2 and spin-1 Kondo effects in a quantum dot’

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1 Details of the peak-fitting procedure for figures 2 and 3 in the main text

For both high and intermediate field regimes, we did several sets of fits to our data, starting with four completely unconstrained Lorentzians at each field (representing two low-bias Kondo peaks and two higher-bias inelastic cotunnelling peaks) on various (parabolic, V-shaped, flat) backgrounds. After each set, we looked at the goodness of the fits through numerical measures as well as by comparing fit results slice-by-slice (field-by-field) to the actual data. Through this procedure we determined that our
background is more V-shaped than parabolic or flat and is very constant through the field range.

We then started pruning the number of free parameters and studying the results as before.

For the high field regime (4-7T) we found that we could obtain very good fits to our data while holding the V-shaped background and the two side peaks fixed — the free parameters here are then the weight, width and location of the central Kondo peaks. We found that adding further constraints — such as requiring the central peaks to be symmetrically spaced about zero bias — worsened the quality of the fits considerably, so we stopped here.

For the lower fields around 3.5T, we also obtained very good fits for fixed V-shaped background and side peaks (with slightly different parameter values for the side peaks as noted in the paper). We next also constrained the central peaks to have equal widths and be equidistant from zero bias. Note that the free parameters here are thus two fewer than before — the weights of the central Kondo peaks, plus the width and distance from zero bias which are the same for both.

The quality of fits remained excellent and these additional constraints did not change the results over a wide field range: 2.3-6T. This is the range for which we report results for this second 'intermediate field' fit.

When we observed the sublinear splitting described in the paper, we considered that temperature-broadening might have the effect of making peaks appear to be closer together than they actually were, especially in the crucial intermediate field regime. We therefore included temperature-broadening of 350mK (which was the measurement temperature) in our fits but found that it had a negligible effect, so we ended up leaving this out.

As can be seen in the figures, our two fitting procedures produce nearly-indistinguishable results over a large overlap region of 4-6T, another indication of the robustness of the fits.
2 Comparing existing calculations to transport experiments

We wish to note some issues in comparing existing calculations to any transport experiment and in particular to ours.

1. All the theories calculate spin-resolved density of states, whereas experiments measure the sum of the two spin-resolved densities of states. We address this by fitting our measurements with two Lorentzian peaks and interpreting the extracted peak positions as the positions of the resonances in the spin-resolved density of states. We find that the results are insensitive to details of the fitting procedure for sufficiently high fields, \( g\mu_B B > 2.3 T(1.6kT_K) \). At lower fields we do not report peak positions, as details of the fitting procedure influence the extracted peak positions when the peaks strongly overlap. Information on the actual non-Lorentzian (and non-analytic) Kondo density of states at small but finite \( B \) would likely be necessary to extract reliable (self-consistent) splittings at these low fields.

2. Lineshape asymmetries at high magnetic field predicted by Rosch et al. and Logan-Dickens may also influence our extracted peak positions [S1, S2] but the fact that we extract splitting that is highly linear with field at high field, extrapolating to zero at zero field, suggests that these issues of lineshape do not have a strong effect on our extracted splittings.

3. Most theories calculate an equilibrium density of states. In transport experiments a finite bias is applied across the quantum dot, changing the density of states of the system. Konik et al. extended the Moore-Wen Bethe-ansatz analysis to take into account non-equilibrium effects, and achieved very similar predictions. [S3] We expect non-equilibrium effects to be small in our experiments since our dot is coupled much more strongly to one lead than to the other: the ratio of couplings is roughly 8 as judged from our maximum observed conductance.
3 The underscreened spin-1 Kondo effect

A quantum dot with spin-1 can only be fully screened by two screening channels in the leads, which we shall label with index \( \alpha = 1, 2 \). Each channel will have its own coupling strength to the dot and an associated Kondo temperature, \( T_{K\alpha} \) \[S4\].

In the case where \( T_{K1} \neq T_{K2} \) and \( T_{K1} \sim T_{K2} \), a two-stage Kondo effect can be observed as reported in Ref.s \[S5\] and \[S6\]. The conductance signature of this configuration is striking: a dip with width corresponding to the smaller of the \( T_{K} \)s is superimposed on the main zero bias peak of width corresponding to the larger of the \( T_{K} \)s. Of course one must have \( T \ll T_{K1}, T_{K2} \) to observe this structure.

In the case where \( T, T_{K1} \ll T_{K2} \), it is possible to have an underscreened Kondo effect — there is only one well-coupled screening channel in the leads and so the total spin of ‘dot and leads’ is reduced only to \( 1/2 \) rather than 0.

The observation of a simple peak in a spin-1 diamond as in our work and others’ \[S7–S9\] is suggestive of such a scenario; however, recently, Posazhennikova and Coleman \[S10\] have proposed further, more striking signatures of the underscreened Kondo effect — according to their noncrossing approximation (NCA) calculations there should be a singularity at low temperature and voltage such that \( d^2I/dVdE \) diverges with increasing \( 1/E \), where \( E \) is either \( V \) or \( T \).

We were unfortunately unable to test this prediction as we did not have enough data points at low energies; however, we wanted to note and call attention to this important theoretical calculation.

4 The finite-bias feature in Figures 4a and c

There is a strongly gate-dependent finite-bias feature in Figure 4. Is this due somehow to the likely encapsulation of buckyballs in our nanotube? Though we cannot exclude the possibility, we believe it is unlikely for the following reasons:

1. Our earlier work and that of other researchers has shown that as a whole \( C_{60} \) peapods look similar to unfilled tubes in transport at low biases. \[S11,S12\]
2. Similar gate-dependent finite-bias Kondo features have previously been observed in unfilled nanotubes. See for example Ref. [S13] As in our case, the mechanism for the gate-dependence is not identified in that work.

3. The energy of the unexplained feature (∼1 – 12meV) is too low to match vibrational states (>30meV) of C₆₀, so it is not clear what C₆₀-associated mechanism would produce the feature. [S14]

We propose a simpler explanation: This feature may mark the transition between the lowest $m_z = 0$ triplet state and the first excited singlet state. This candidate transition does not change $m_z$, and hence does not move with magnetic field, in agreement with observation. We do not know the mechanism for gate-tuning of this feature nor whether the fact that it appears to pass through one corner of the diamond is anything more than an accidental alignment of energy levels at that point.

References


