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Three-body correlations in nonlinear response of correlated quantum liquid

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Understanding the properties of correlated quantum liquids is a fundamental issue of condensed matter physics. Even in such a correlated case, fascinatingly, we can tell that the equilibrium fluctuations of the system govern its linear response to an external field, relying on the fluctuation dissipation relations based on the two-body correlations. Going beyond, up to the three-body correlations, is of importance for van der Waals force [1], the three-body force in nuclei [2], the Efimov state [3, 4], the ring exchange interaction in solid ³He [5, 6], and frustrated spin systems [7]. In our work, we have used a quantum dot in the Kondo regime, which is a controllable realization of such a correlated quantum liquid [8–11]. Thanks to the quality of our sample, where the Kondo effect in the unitary limit was achieved, we could quantitatively measure the three-body correlations and their role in the non-equilibrium regime, in perfect agreement with recent results of the Fermi liquid theory [12–15]. In particular, we have demonstrated its importance when time-reversal symmetry is broken, solving a long-standing puzzle of the Kondo systems under the magnetic field [13]. The demonstrated method to relate three-body correlation and non-equilibrium transport opens up a way for further investigation of the dynamics of quantum many-body systems.

A quantum dot (QD) system enables us to study the Kondo effect, i.e., the formation of a spin singlet state between the impurity spin and conduction electrons in the reservoirs, with high controllability and accessibility to the spin degree of freedom [8–11]. The essential physics can be captured by the Anderson impurity model, which describes the single impurity state with a spin σ ($=\uparrow, \downarrow$ or ± 1) and energy ε_σ coupled to the two metallic reservoirs. The Fermi liquid theory for the Anderson model, which has been developed phenomenologically [16] and microscopically [17–21], can explain the low-energy physics of highly correlated quantum systems as the properties of

an ensemble of quasi-particles interacting with each other via residual interaction. The linear response at zero magnetic field, namely when the time-reversal symmetry is present, can be explained in terms of only three parameters; the phase shift δ_σ when by a spin passes through the impurity state, and the two-body correlations ($\chi_{\sigma_1\sigma_2}$) between two different spins (σ_1 and σ_2). These parameters will determine the two crucial physical quantities of the Kondo problem: the Kondo temperature $T_K \equiv 1/(4\chi_{\uparrow\uparrow})$ and the Wilson ratio $R \equiv 1 - \chi_{\uparrow\downarrow}/\chi_{\uparrow\uparrow}$ [16, 17].

To go beyond this well-established regime, nonlinear susceptibility, that is, three-body correlation $\chi_{\sigma_1\sigma_2\sigma_3}$ comes into the game. Consider a situation where three electrons with spin σ_1 , σ_2 , and σ_3 pass through the QD level in sequence as schematically shown in Fig. 1 (a). The electron with σ_1 first occupies and leaves the level, and then the second electron with σ_2 passes through it. Finally, the third electron with σ_3 occupies it. This process describes that the spin in the QD level fluctuates from σ_1 to σ_2 to σ_3 [from $\uparrow$ to $\uparrow$ to $\downarrow$, in the specific case shown in Fig. 1(a)] and that the spin of the first electron affects that of the third spin. The correlation among σ_1 and σ_3 via σ_2 , or three-spin exchange process, gives the most naive picture of the three-body correlation, which constitutes the leading term of the nonlinear response of the system as characterized by $\chi_{\sigma_1\sigma_2\sigma_3}$ ($\chi_{\uparrow\uparrow\downarrow}$ in this specific case). Note that the theoretical definition of the three-body correlation is the differential of the free energy Ω of the total system regarding the energies of those three electrons, namely $\chi_{\sigma_1\sigma_2\sigma_3} \equiv -\partial^3\Omega/\partial\varepsilon_{\sigma_1}\partial\varepsilon_{\sigma_2}\partial\varepsilon_{\sigma_3}$, (see Supplementary Information).

In this paper, we experimentally prove that $\chi_{\sigma_1\sigma_2\sigma_3}$ indeed contributes to the nonlinear conductance. We study the differential conductance (dI/dV) of the Kondo-correlated QD with the following expansion in terms of the bias voltage (V) [22–28]:

$$\frac{dI}{dV} = G_0 - \alpha_V \left(\frac{eV}{k_B T_K} \right)^2 + \cdots, \quad (1)$$

at $T \ll T_K$. Here G_0 , e , k_B , and T_K , are the zero-bias conductance, electron charge, the Boltzmann constant,

FIG. 1: (a) Schematic view of the three-body correlation $\chi_{\uparrow\uparrow\downarrow}$. Electrons fluctuate between the dot and the leads in the equilibrium and all those processes are summed up in $\chi_{\uparrow\uparrow\downarrow}$. (b) (center) Quantum dot (QD) at time-reversal symmetry (TRS) and particle-hole symmetry (PHS) points. The energy level is $\varepsilon_\sigma = -U/2$, where U is Coulomb interaction. The two-body correlations $\chi_{\sigma_1\sigma_2}$ is finite, while the three-body correlation is quenched. (left) QD with broken TRS, where the two spins are separated by $2\mu_B B$. (right) QD with broken PHS, where the energy level is $\varepsilon_\sigma \neq -U/2$. Experimentally, the gate voltage is applied to QD in order to tune the energy level. In the broken symmetry cases, the three-body correlations $\chi_{\sigma_1\sigma_2\sigma_3}$ is finite, which we detect in this paper.

and the Kondo temperature at zero magnetic field. The coefficient α_V of the order V^2 consists of two- and three-body terms, W_2 and W_3 . W_2 is defined using δ_σ and $\chi_{\sigma_1\sigma_2}$, which was addressed and established experimentally [29–33]. Here, we newly focus on W_3 , which is given by δ_σ , $\chi_{\uparrow\uparrow}$, and $\chi_{\sigma_1\sigma_2\sigma_3}$ (see Supplementary Information for the precise formula) [12–15]. We rely on the fact that $G_0 = e^2/h(\sin^2 \delta_\uparrow + \sin^2 \delta_\downarrow)$ in the Kondo effect in the unitary limit. Such a treatment is possible in this experimental work as we have achieved the unitary limit in our clean carbon nanotube QD [33, 34].

In the $U = 0$ case, the complete analytical form for Eq. (1) is obtained, where U is the charging energy of QD (see Supplementary Information). Note that the three-body correlations for the same spins ($\chi_{\uparrow\uparrow\uparrow}$ and $\chi_{\downarrow\downarrow\downarrow}$) are finite even in the $U = 0$ case due to the Pauli exclusion principle. On the other hand, the Kondo effect makes other components such as $\chi_{\uparrow\uparrow\downarrow}$ finite. Thus, the experimental observation of the deviation from the $U = 0$ case directly tells us the relevance of those correlations in the nonlinear Kondo regime. In addition, a theory recently reported α_V in the interacting case by expanding the Fermi liquid theory to the non-equilibrium regime [12–15]. Our present work, relying on a very precise experiment, constitutes an accurate test of this theory.

We perform conductance measurement for a carbon nanotube connected to two Pd/Al electrodes in the dilution fridge, using a standard low-frequency lockin technique [33–36]. The base temperature of the dilution fridge is 16 mK. A small magnetic field 0.1 T, which is much smaller compared to the Kondo temperature ($T_K = 1.6$ K), is always applied to suppress the superconductivity of the electrodes. In this paper, for simplicity, we refer to this lowest field 0.1 T as zero field (0 T), except when we mark experimental points at $B = 0.1$ T in figures.

FIG. 2: (a) ε/U dependence of G_0 for three temperatures (16, 380, and 780 mK). The experimental T_K^* shown on the right axis is fitted with the formula shown in Ref. [33]. (b) dI/dV as a function of $eV/k_B T_K$ for different magnetic fields from 0 to 2.5 T with 0.1 T steps. Here, the lowest field 0.1 T is defined as 0 T. (inset) 2Δ , which is the width of Zeeman splitting, as a function of $\mu_B B/k_B T_K$.

A single electron energy with spin σ in the QD is defined as $\varepsilon_\sigma = \varepsilon - g\sigma\mu_B B/2$, where μ_B and $g \approx 2$ are the Bohr magneton and the g -factor in the nanotube, respectively. In the time-reversal symmetric (TRS) and particle-hole symmetric (PHS) case, the energy levels are at $\varepsilon_\sigma = -U/2$ [center of Fig. 1(b)], which many experimental works have addressed [29–33]. In the symmetrical points, the two-body correlation is finite, while the three-body correlation is quenched due to the symmetry. We tune ε_σ to investigate the behavior of the coefficient α_V by applying either magnetic field (B) or gate voltage (V_g). As is well known in the Kondo physics, the magnetic field splits the degenerate level with the splitting width $2\mu_B B$, which breaks TRS [left of Fig. 1(b)]. Gate voltage, on the other hand, moves ε from $-U/2$ and breaks PHS [right of Fig. 1(b)] [12–15].

Figure 2(a) shows the zero-bias conductance (G_0) as a function of ε/U at zero field for three different temperatures. $\varepsilon/U = -0.5$ corresponds to the PHS point. In this paper, we define T_K as the Kondo temperature at the TRS and the PHS point: $B = 0$ T and $\varepsilon/U = -0.5$. We define the Kondo temperature $T_K^*(\varepsilon, B)$ for broken symmetry cases. $T_K^*(\varepsilon/U = -0.5, B = 0 \text{ T}) \equiv T_K = 1.6$ K. The black points in Fig. 2(a) are the Kondo temperatures $T_K^*(\varepsilon, B = 0 \text{ T})$, which we evaluate by analyzing the temperature dependence of G_0 [33]. We obtain $U = 6.4 \pm 0.8$ meV and $\Gamma = 1.9 \pm 0.2$ meV by fitting the gate voltage dependence of $T_K^*(\varepsilon, B = 0 \text{ T})$ (dashed curve), where Γ is the width of energy levels due to coupling between the electron in the QD and the lead electrons [33]. $R = 1.95 \pm 0.05$ which we independently determined from the shot noise experiment [33] together with $U/\Gamma = 3.4 \pm 0.6$ indicates that the Kondo state is very close to the limit of the strong correlation regime [23]. As the two-body correlation W_2 is defined [14, 15] by:

$$W_2 = - \left[1 + 5(R - 1)^2 \right] \sum_{\sigma} \frac{\cos 2\delta_{\sigma}}{2}, \quad (2)$$

$W_2 = 6.0$ at the symmetric point $\delta_{\sigma} = \pi/2$. In the following, we investigate the effect of the symmetry breaking of TRS [Fig. 2(b) and Fig. 3] and PHS [Fig. 4].

The differential conductance was obtained at 16 mK and PHS point ($\varepsilon/U = -0.5$) as a function of source-drain voltage V for different B from 0 to 2.5 T [Fig. 2(b)]. When B increases, the Kondo resonance splits, and its amplitude is reduced. Inset in Fig. 2(b) shows the Zeeman splitting 2Δ as a function of $\mu_B B/k_B T_K$. The linear fitting shows that the g -factor is 2.0 ± 0.05 just as expected for carbon nanotube and the splitting seems to start at $\mu_B B/k_B T_K = 0.23 \pm 0.02$ ($B = 0.6$ T). Although this splitting has served as a hallmark of the Kondo effect [8–11], the microscopic mechanism has been theoretically revealed only recently [13–15].

We analyze the magnetic field dependence of the Kondo peak with Eq. (1) (the analysis procedure

FIG. 3: (a) W_2 and m_d as a function of $\mu_B B/k_B T_K$. The points are experimental results, while the lines are theoretical ones. The dotted and dashed lines for W_2 are given by the free particle (FP) model and the NRG calculations ($U/\Gamma = 3.5$), respectively. (b) α_V as a function of $\mu_B B/k_B T_K$. (Inset) dI/dV as a function of $\text{sign}(V) \times (eV/k_B T_K)^2$ at different magnetic fields. The curves are offset for clear comparison. We obtained α_V with linear fitting. (c) Three-body correlations W_3 as a function of $\mu_B B/k_B T_K$. In the analysis, we have compared the experimental results with the FP model (dotted lines) and the exact NRG results for the Anderson model with $U/\Gamma = 3.5$ (dashed lines) [14, 15].

can be found in the Supplementary Information). At a finite magnetic field, the phase shift relates to the induced magnetization of the electron inside the QD m_d : $m_d = (\delta_{\uparrow} - \delta_{\downarrow})/\pi$, with which G_0 and W_2 are expressed as $G_0 = \frac{2e^2}{h} \cos^2(\pi m_d/2)$ and $W_2 = - \left[1 + 5(R - 1)^2 \right] \cos(\pi m_d)$, respectively [14]. Note that to derive this we use the Friedel sum rule, which

holds even under the magnetic field. We show the gradual increase of m_d deduced from the experimental G_0 shown on the right axis in Fig. 3(a). The numerical renormalization group (NRG) calculation tells that the Wilson ratio is almost insensitive to the magnetic field up to $\mu_B B/k_B T_K = 1$ (see Supplementary Information). Thus, we can directly deduce the field dependence of the two-body correlation W_2 with the experimental value of the Wilson ratio $R = 1.95$ at zero field [33] and of m_d [Fig. 3(a)]. To the best of our knowledge, a quantitative measurement of the two-body correlations of the Kondo state under magnetic fields has never been reported.

Let us now analyze the non-equilibrium case of Fig. 2(b) in more detail based on Eq. (1). The inset of Fig. 3(b) shows dI/dV as a function of $\text{sign}(V) \times (eV/k_B T_K)^2$ at $B = 0, 0.5, 1, 1.5$, and 2.5 T, respectively. The “V-shaped” curves indicate that the conductance shows a parabolic behavior around zero bias, and importantly, the sign of their curvature changes around 1 T when B increases. This sign reversal can also define the splitting of the Kondo peak, on which we focus here, instead of the above 2Δ [37]. We obtain α_V from the linear fitting with the second term of Eq. (1) [38, 39]. Figure 3(b) is the curvature α_V as a function of the rescaled magnetic field $\mu_B B/k_B T_K$. α_V crosses zero at $\mu_B B/k_B T_K = 0.38$ ($B = 0.9$ T).

In order to elucidate the physical meaning of the above magnetic field dependence of α_V , we first consider the free particle (FP) model with a resonant level and width (see Supplementary Information). In this case, analytic solutions of α_V are given only by using a single parameter, coupling constant ($2\Gamma_0$), corresponding to the half width of dI/dV at zero field. When Γ_0 is given, the differential conductance in the magnetic field can be calculated for the FP case. By assuming $\Gamma_0 = \Gamma$, $\alpha_V(B)$ thus obtained is shown in the dotted curve in Fig. 3(b). The dotted curve fails to explain the experimental observation, indicating the relevance of the Kondo correlation. Now, we carry out NRG calculation, which gives $\alpha_V(B)$ for the Kondo-correlated QD with $U/\Gamma = 3.5$ (dashed line) [14]. The theoretical curve satisfactorily reproduces the experiment, as shown in Fig. 3(b).

Then what can we learn from this agreement between the experiment and the theory? As recently discussed [13–15], α_V is related to W_2 and W_3 :

$$\alpha_V(B) = \frac{2e^2}{h} \frac{\pi^2}{64} (W_2 + W_3) \times \left(\frac{T_K}{T_K^*} \right)^2. \quad (3)$$

Here, W_3 is defined as $W_3 = \frac{1}{2} \sum_{\sigma} \frac{\sin 2\delta_{\sigma}}{2\pi} \times (\chi_{\sigma\sigma\sigma} + 3\chi_{\sigma-\sigma-\sigma})/\chi_{\uparrow\uparrow}^2$. As explained, we have already obtained the magnetic field dependence of W_2 . $T_K^*(\varepsilon = 0, B)$ as a function of B is obtained from the Lorentzian fitting of dI/dV (see Supplementary Information). In this way, we finally evaluate the three-body correlations (W_3) with Eq. (3) [Fig. 3(c)]. W_3 is zero at zero field

FIG. 4: (a) G_0 and δ as a function of ε/U . The points are experimental results. The dashed line for G_0 is given by the NRG calculations with $U/\Gamma = 3.5$. (b) α_V as a function of ε/U . (Inset) dI/dV as a function of $\text{sign}(V) \times (eV/k_B T_K)^2$ at $\varepsilon/U = -0.51$ and -0.65 . (c) Three-body correlations W_3 as a function of ε/U . We compare the experimental results with the NRG results with $U/\Gamma = 3.5$ (dashed lines) [14, 15].

and becomes finite as the magnetic field increases i.e., as the TRS is broken. Importantly, the absolute value of the experimental W_3 is much larger than the theoretical curve by the FP model. The behavior of W_3 is due to the enhancement of three-body correlations $\chi_{\sigma_1\sigma_2\sigma_3}$ by the Kondo effect. Indeed, NRG calculations with $U/\Gamma = 3.5$ shown in a dashed curve nicely reproduce the experimental W_3 [see also the comparison of W_2 in Fig. 3(a)]. This result shows that the residual interaction manifests itself in the non-equilibrium transport in the symmetry-breaking regime. The experimental determination of the three-body correlation is our central achievement of this work.

So far, we have focused on the PHS point [see Fig. 1(b)]. Next, we address the PHS breaking regime by tuning gate voltage (namely, ε). The square mark in Fig. 4(a) shows ε/U dependence of the G_0 , which decreases as ε deviates from the symmetry point ($\varepsilon/U = -0.5$). The dashed line given by the NRG calculation again agrees with the experimental points well. We also show the phase shift, $\delta_\sigma (= \delta_\uparrow = \delta_\downarrow)$, evaluated from Eq. (1) [circles in Fig. 4(a)]. As expected, the phase is locked around $\pi/2$ and develops a plateau [40, 41]. The NRG calculation shows that the Wilson ratio does not decrease in this region (see Supplementary Information). Thus, we evaluate ε/U dependence of the two-body correlation W_2 with Eq. (2) and the experimental value of the Wilson ratio $R = 1.95$ at zero field [see Fig. S5 in Supplementary Information].

Inset of Fig. 4 (b) shows dI/dV with offset as a function of $\text{sign}(V) \times (eV/k_B T_K)^2$ at $\varepsilon/U = -0.51$ and -0.65 . The slope at the former (PHS point) is larger than the latter (PHS broken). Figure 4(b) is the deduced curvature of the differential conductance α_V as a function of ε/U . Here, we investigate the region within $|\varepsilon/U + 0.5| < 0.15$, where we experimentally obtain the Kondo temperature, $T_K^*(\varepsilon, B = 0 \text{ T})$ [see Fig. 2(a)]. In Fig. 4(c), we plot W_3 evaluated with Eq. (3), showing that W_3 remains almost zero in a wide region $|\varepsilon/U + 0.5| < 0.15$, which is in sharp contrast with the above result in the magnetic field. This indicates that three-body correlation is more sensitive to magnetic field modulation than the gate voltage modulation (energy level modulation), which reflects that the spin degree of freedom of the Kondo state is fluctuating while the charge part is frozen. Again, the NRG calculation with $U/\Gamma = 3.5$ shown in a dashed curve agrees with experimental α_V and W_3 . This agreement clearly tells that the three-body correlation can be estimated experimentally.

In conclusion, we report the experimental determination of non-equilibrium many-body correlations in the Kondo QD. We demonstrate that the curvature of the differential conductance in the TRS and PHS breaking regimes is reproduced well by the NRG calculation with strong interactions, enabling us to successfully evaluate the two-body and three-body correlations. The quantitative analysis strongly confirmed the recent development of the Fermi liquid theory in non-equilibrium situations, which should inspire future work. In particular, sensitivity to TRS and PHS should make three-body correlations an efficient tool to explore correlated quantum liquids such as topological spin liquids.

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- [1] Axilrod, B. M. & Teller, E. Interaction of the van der Waals type between three atoms. *The Journal of Chemical Physics* **11**, 299–300 (1943).
 - [2] Otsuka, T., Suzuki, T., Holt, J. D., Schwenk, A. & Akaishi, Y. Three-body forces and the limit of oxygen isotopes. *Phys. Rev. Lett.* **105**, 032501 (2010).
 - [3] Efimov, V. Energy levels arising from resonant two-body forces in a three-body system. *Physics Letters B* **33**, 563–564 (1970).
 - [4] Kraemer, T. *et al.* Evidence for Efimov quantum states in an ultracold gas of caesium atoms. *Nature* **440**, 315–318 (2006).
 - [5] Thouless, D. J. Exchange in solid ^3He and the Heisenberg Hamiltonian. *Proceedings of the Physical Society* **86**, 893–904 (1965).
 - [6] Roger, M., Hetherington, J. H. & Delrieu, J. M. Magnetism in solid ^3He . *Rev. Mod. Phys.* **55**, 1–64 (1983).
 - [7] Lacroix, C., Mendels, P. & Mila, F. *Introduction to Frustrated Magnetism: Materials, Experiments, Theory*. Springer Series in Solid-State Sciences (Springer Berlin Heidelberg, 2011). URL <https://books.google.co.jp/books?id=utSV09Zuh0kC>.
 - [8] Goldhaber-Gordon, D. *et al.* Kondo effect in a single-electron transistor. *Nature* **391**, 156–159 (1998).
 - [9] Cronenwett, S. M., Oosterkamp, T. H. & Kouwenhoven, L. P. A tunable Kondo effect in quantum dots. *Science* **281**, 540–544 (1998).
 - [10] Schmid, J., Weis, J., Eberl, K. & Klitzing, K. v. A quantum dot in the limit of strong coupling to reservoirs. *Physica B: Condensed Matter* **256–258**, 182–185 (1998).
 - [11] van der Wiel, W. G. *et al.* The Kondo effect in the unitary limit. *Science* **289**, 2105–8 (2000).
 - [12] Mora, C., Moca, C. P., von Delft, J. & Zarand, G. Fermi-liquid theory for the single-impurity Anderson model. *Physical Review B* **92**, 075120 (2015).
 - [13] Filippone, M., Moca, C. P., Weichselbaum, A., von Delft, J. & Mora, C. At which magnetic field, exactly, does the Kondo resonance begin to split? A Fermi liquid description of the low-energy properties of the Anderson model. *Phys. Rev. B* **98**, 075404 (2018).
 - [14] Oguri, A. & Hewson, A. C. Higher-order Fermi-liquid corrections for an Anderson impurity away from half filling: Nonequilibrium transport. *Physical Review B* **97**, 035435 (2018).
 - [15] Oguri, A. & Hewson, A. C. Higher-order Fermi-liquid corrections for an Anderson impurity away from half filling. *Physical Review Letters* **120**, 126802 (2018).
 - [16] Nozieres, P. A "Fermi-liquid" description of the Kondo problem at low temperatures. *Journal of Low Temperature Physics* **17**, 31–42 (1974).
 - [17] Yosida, K. & Yamada, K. Perturbation expansion for the Anderson Hamiltonian. *Progress of Theoretical Physics Supplement* **46**, 244–255 (1970).
 - [18] Yamada, K. Perturbation expansion for the Anderson Hamiltonian. II. *Progress of Theoretical Physics* **53**, 970–986 (1975).
 - [19] Yamada, K. Perturbation expansion for the Anderson Hamiltonian. IV. *Progress of Theoretical Physics* **54**, 316–324 (1975).
 - [20] Shiba, H. The Korringa relation for the impurity nuclear spin-lattice relaxation in dilute Kondo alloys. *Progress of*

- Theoretical Physics* **54**, 967–981 (1975).
- [21] Yoshimori, A. Perturbation analysis on orbital-degenerate Anderson model. *Progress of Theoretical Physics* **55**, 67–80 (1976).
 - [22] Pustilnik, M. & Glazman, L. Kondo effect in quantum dots. *Journal of Physics: Condensed Matter* **16**, R513–R537 (2004).
 - [23] Oguri, A. Fermi liquid theory for the nonequilibrium Kondo effect at low bias voltages. *Journal of the Physical Society of Japan* **74**, 110–117 (2005).
 - [24] Rincon, J., Aligia, A. A. & Hallberg, K. Universal scaling in nonequilibrium transport through an Anderson impurity. *Physical Review B* **79**, 121301 (2009).
 - [25] Rincon, J., Aligia, A. A. & Hallberg, K. Erratum: Universal scaling in nonequilibrium transport through an Anderson impurity. *Physical Review B* **81**, 039901 (2010).
 - [26] Roura-Bas, P. Universal scaling in transport out of equilibrium through a single quantum dot using the noncrossing approximation. *Physical Review B* **81**, 155327 (2010).
 - [27] Sela, E. & Malecki, J. Nonequilibrium conductance of asymmetric nanodevices in the Kondo regime. *Physical Review B* **80** (2009).
 - [28] Mora, C., Vitushinsky, P., Leyronas, X., Clerk, A. A. & Le Hur, K. Theory of nonequilibrium transport in the SU(N) Kondo regime. *Physical Review B* **80**, 155322 (2009).
 - [29] Grobis, M., Rau, I. G., Potok, R. M., Shtrikman, H. & Goldhaber-Gordon, D. Universal scaling in nonequilibrium transport through a single channel Kondo dot. *Phys Rev Lett* **100**, 246601 (2008).
 - [30] Scott, G. D., Keane, Z. K., Cizek, J. W., Tour, J. M. & Natelson, D. Universal scaling of nonequilibrium transport in the Kondo regime of single molecule devices. *Physical Review B* **79**, 165413 (2009).
 - [31] Yamauchi, Y. *et al.* Evolution of the Kondo effect in a quantum dot probed by shot noise. *Physical Review Letters* **106**, 176601 (2011).
 - [32] Kretinin, A. V. *et al.* Spin- $\frac{1}{2}$ Kondo effect in an InAs nanowire quantum dot: Unitary limit, conductance scaling, and Zeeman splitting. *Physical Review B* **84**, 245316 (2011).
 - [33] Ferrier, M. *et al.* Universality of non-equilibrium fluctuations in strongly correlated quantum liquids. *Nature Physics* **12**, 230–235 (2016).
 - [34] Ferrier, M. *et al.* Quantum fluctuations along symmetry crossover in a Kondo-correlated quantum dot. *Physical Review Letters* **118**, 196803 (2017).
 - [35] Teratani, Y. *et al.* Field-enhanced Kondo correlations in a half-filling nanotube dot: Evolution of an SU(N) Fermi-liquid fixed point. *Journal of the Physical Society of Japan* **85**, 094718 (2016).
 - [36] Hata, T. *et al.* Enhanced shot noise of multiple Andreev reflections in a carbon nanotube quantum dot in SU(2) and SU(4) Kondo regimes. *Phys. Rev. Lett.* **121**, 247703 (2018).
 - [37] Hewson, A. C., Bauer, J. & Oguri, A. Non-equilibrium differential conductance through a quantum dot in a magnetic field. *Journal of Physics: Condensed Matter* **17**, 5413 (2005).
 - [38] In this analysis, we do not take the data obtained at very low bias region (typically $|eV/k_B T_K|^2 < 0.01$), where a slight deviation from the parabolicity is observed due to some other lower energy physics such as two-stage Kondo effect (see Supplement and the reference below).
 - [39] van der Wiel, W. G. *et al.* Two-stage Kondo effect in a quantum dot at a high magnetic field. *Physical Review Letters* **88**, 126803 (2002).
 - [40] Takada, S. *et al.* Transmission phase in the Kondo regime revealed in a two-path interferometer. *Phys. Rev. Lett.* **113**, 126601 (2014).
 - [41] Takada, S. *et al.* Low-temperature behavior of transmission phase shift across a Kondo correlated quantum dot. *Phys. Rev. B* **94**, 081303 (2016).

Figures

Figure 1

(a) Schematic view of the three-body correlation $\chi_{\sigma_1\sigma_2\sigma_3}$. Electrons fluctuate between the dot and the leads in the equilibrium and all those processes are summed up in $\chi_{\sigma_1\sigma_2\sigma_3}$. (b) (center) Quantum dot (QD) at time-reversal symmetry (TRS) and particle-hole symmetry (PHS) points. The energy level is $\varepsilon_\sigma = -U/2$, where U is Coulomb interaction. The two-body correlations $\chi_{\sigma_1\sigma_2}$ is finite, while the three-body correlation is quenched. (left) QD with broken TRS, where the two spins are separated by $2\mu_B B$. (right) QD with broken PHS, where the energy level is $\varepsilon_\sigma \neq -U/2$. Experimentally, the gate voltage is applied to QD in order to tune the energy level. In the broken symmetry cases, the three-body correlations $\chi_{\sigma_1\sigma_2\sigma_3}$ is finite, which we detect in this paper

Figure 2

(a) ϵ/U dependence of G_0 for three temperatures (16, 380, and 780 mK). The experimental T_K (K) shown on the right axis is fitted with the formula shown in Ref. [33]. (b) dI/dV as a function of $eV/k_B T_K$ for different magnetic fields from 0 to 2.5 T with 0.1 T steps. Here, the lowest field 0.1 T is defined as 0 T. (inset) 2Δ , which is the width of Zeeman splitting, as a function of $\mu_B B / k_B T_K$.

Figure 3

(a) W_2 and m_d as a function of $\mu_B B / k_B T_K$. The points are experimental results, while the lines are theoretical ones. The dotted and dashed lines for W_2 are given by the free particle (FP) model and the NRG calculations ($U/\Gamma = 3.5$), respectively. (b) α_V as a function of $\mu_B B / k_B T_K$. (Inset) dI/dV as a function of $\text{sign}(V) \times (eV/k_B T_K)^2$ at different magnetic fields. The curves are offset for clear comparison. We obtained α_V with linear fitting. (c) Three-body correlations W_3 as a function of $\mu_B B / k_B T_K$. In the analysis, we have compared the experimental results with the FP model (dotted lines) and the exact NRG results for the Anderson model with $U/\Gamma = 3.5$ (dashed lines) [14, 15].

Figure 4

(a) G_0 and δ as a function of ϵ/U . The points are experimental results. The dashed line for G_0 is given by the NRG calculations with $U/\Gamma = 3.5$. (b) α_V as a function of ϵ/U . (Inset) dI/dV as a function of $\text{sign}(V) \times (eV/k_B T_K)^2$ at $\epsilon/U = -0.51$ and -0.65 . (c) Three-body correlations W_3 as a function of ϵ/U . We compare the experimental results with the NRG results with $U/\Gamma = 3.5$ (dashed lines) [14, 15].

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