Low Temperature Physics/Fizika Nizkikh Temperatur, 2013, v. 39, No. 3, pp. 335–342

Switchable zero-bias anomaly in individual C_{60} molecules contacted with tunable aluminum electrodes

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Received November 7, 2012

We report the observation of strong resonances at zero bias in the differential conductance through Al–C_{60}–Al junctions with tunable electrode distance, measured above $T = 10$ K. The conductance value at resonance ranges from a few percent up to eighty percent of the quantum of conductance. The resonances may disappear or reoccur completely and discontinuously upon very small changes of the electrode distance. However, once they are formed they are very robust with respect to changes of the electrode distance. We discuss similarities and differences to the common theories of the Kondo screening of a spontaneous spin polarization of the C_{60} molecule. We deduce Kondo temperatures in the range from 35 to 160 K and demonstrate that the temperature dependence is in agreement with the scaling behavior of the Kondo effect in the temperature range of our experiment.

PACS: **73.63.–b** Electronic transport in nanoscale materials and structures;
73.23.–b Electronic transport in mesoscopic systems;
74.70.Wz Carbon-based superconductors;
85.65.+h Molecular electronic devices.

Keywords: Kondo effect, fullerenes, point-contact spectroscopy, break-junctions.

1. Introduction

In a metal with magnetic impurities, the individual localized impurity spins can effectively be shielded by the conduction electrons, which gives rise to enhanced resistance at low temperatures. This phenomenon is called Kondo effect and has been thoroughly studied in the 1960s to 1980s in bulk systems [1]. Its signature is a resonance in the density of states located at the Fermi energy. The width of the resonance is given by the Kondo temperature $k_B T_K$. Because of its magnetic origin, the resonance is splitting in an external magnetic field according to the Zeeman energy. The size of T_K itself depends on the magnetic species and the host metal and can vary between a few millikelvin and several hundred Kelvin [1]. In 1998 Goldhaber–Gordon and coworkers discovered enhanced zero-bias conductance in the transport through semiconductor quantum dots, when they were charged with an odd number of extra electrons and their coupling to the electrodes was in the so-called intermediate range [2,3]. The term intermediate regime refer to a situation, in which the probability of simultaneous transport of charges across the barriers of the island is not negligible, but the coupling is weak enough to consider the island as a separate subunit of the circuit on which, e.g., the electrostatic potential is well-defined. As a rule of thumb the intermediate regime is realized when with symmetric coupling of the island to both leads, the high-bias conductance is in the range of 0.01 to 0.1 G_0. Despite its opposite sign compared to the aforementioned phenomenon, it has soon been understood to be caused by the same Kondo screening. Here, the extra electron on the dot interacts with the conduction electrons on both the two leads [4]. This mesoscopic variant of the Kondo effect has been observed in several realizations of quantum dots [2,3], including carbon nanotubes [5,6] and fullerenes [7–9], point contacts [10,11] down to the size of individual atoms

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[12–15], metal clusters trapped in a nanogap [16] and complex organic molecules [17,18]. Recently also the interaction between two individual Kondo impurities [19] as well as the influence of ferromagnetic electrodes [20] have been studied. The typical features, which are in common in all these examples, are a zero bias anomaly (ZBA) with an amplitude and full width at half maximum (FWHM) that varies with temperature and saturates well below the Kondo temperature:

$$T_K = \frac{\sqrt{\Gamma U}}{2} e^{\pi\varepsilon_0(\varepsilon_0+U)/\Gamma U} \approx e^{\pi\varepsilon_0/\Gamma} . \quad (1)$$

This latter approximation holds if the Coulomb energy U necessary for double charging the quantum dot is large compared to the coupling constant Γ and the energy distance ε_0 between the quantum dot level and the Fermi energy. Since no theory exists so far that describes the full functional shape of the resonance for arbitrary coupling, it is often approximated by a Lorentzian with width

$$\mathrm{FWHM}(T) = 2k_B T_{K,\mathrm{eff}} / e , \quad (2)$$

where k_B is the Boltzmann constant and $T_{K,\mathrm{eff}}$ is an effective temperature exceeding the Kondo temperature. Below T_K the FWHM is expected to saturate [2]. Since the Kondo effect is a many body correlation effect between a localized spin and the spins of the conduction electrodes on both sides of the junction, it only occurs for intermediate values of the coupling constant Γ. In many experiments (e.g., in Refs. 2, 3, 7, 9, 24) also the expected universal temperature dependence [21] was shown,

$$G_{\max}(T) = G_{0K}\left[1+\frac{T^2}{T_K^2}(2^{1/s}-1)\right]^{-s} + G_{\mathrm{or}} , \quad (3)$$

where G_{0K} is the height of the Kondo contribution to the peak conductance at zero temperature, $s = 0.22$ [21] and G_{or} is the off-resonance conductance, i.e., the conductance of the junction at voltages outside the resonance peak. In experiments, in which the coupling was relatively weak, such that charging effects were also observable, it could be demonstrated that the Kondo effect only occurred for odd numbers of extra electrons on the dot and thus an unpaired spin 1/2 [2,3]. In the case of stronger coupling, when no charging effects are observed, or when no gate electrode is available, the number of electrons on the dot cannot be determined independently from the transport data. Nevertheless, a ZBA may occur and is often interpreted as Kondo effect [7,16,17,22,23] because of the phenomenology described above. For molecules such as fullerenes that do not provide an unpaired spin in their ground state, it thus remains open, which spin would be responsible for the Kondo effect. It has also been pointed out recently that not all ZBAs which at the first glimpse resemble Kondo resonances are indeed caused by the Kondo effect, since they do not fulfill all predictions of the Kondo effect theories [24]. In particular in the situation when the off-resonance conductance outside the energy range of the ZBA is relatively high $G_{\mathrm{or}} \gtrsim 0.1\, G_0$ alternative interpretations have to be considered [16]. On the other hand many ZBAs observed in point contacts that remained unexplained at the time of their observation have now been re-discussed in the framework of Kondo physics due to the presence of a spontaneous spin polarization [25].

In a simplified model the quantities which determine T_K in this mesoscopic Kondo effect are the energy ε_0 of the topmost filled quantum dot level and the coupling strength of the dot to the electrodes [4]. In Ref. 7 Parks *et al.* report the possibility to continuously tune the Kondo temperature of C_{60} in a rather wide range, when contacted with adjustable Au electrodes [26]. The authors also mention the possibility of abrupt changes of the Kondo features without going into details of this observation. This is the starting point of our work. In this article we report on a study of the transport through individual C_{60} molecules contacted with the help of mechanically controllable break junctions (MCBJs) [27] made of Al. In contrast to Ref. 7 we were not able to continuously and significantly tune the amplitude and the height of the ZBA by changing the mechanical coupling. However, occasionally, the resonance may abruptly disappear and reappear upon very tiny changes of the coupling. We argue that the sudden disappearance/reappearance is caused by charging and discharging the fullerene with one extra electron. We argue that the strong electronegativity of C_{60} [28,29] in conjunction with the covalent/ionic bonding between the fullerene and Al [30] facilitates charge transfer between the electrodes and the molecule.

2. Experimental

The adjustable electrodes consist of lithographically fabricated MCBJs [27]. A 100 nm thick Al layer on a spring steel substrate is used as electrode material. The Al is electrically isolated from the substrate by a 1.5 μm thick polyimide layer. After establishing electrical contacts to the electrode device it is mounted into the breaking mechanism, transferred into a ultra high vacuum chamber. The electrodes are opened for the first time at a pressure $p < 10^{-8}$ Pa and a temperature $T \approx 10$ K. This procedure guarantees clean metal surfaces and thus enables us to study atomic-sized metal contacts as well as molecular junctions in which the molecule is in direct contact to the metal electrodes without contaminants. After the characterization measurements of the electrodes (see next section) the fullerene molecules are evaporated onto the electrodes while the MCBJ is kept at low temperature $T < 15$ K. The amount of the deposited molecules is monitored via a quartz sensor with a precision to 0.01 monolayers (ML). More details about the setup and its calibration and the characterization measurements are described in Refs. 31 and 32.

A source-measure unit, i.e., an instrument that simultaneously feeds a voltage (in our case of 10 to 100 mV) and measures the resulting current, is used for measuring the linear conductance. The measurements of the differential conductance (dI/dV) were performed with a standard lock-in amplifier. The signals were pre-amplified by a variable gain preamplifier allowing the recording of dI/dV's in a range of kΩ to GΩ. About 10 to 15% of the contacts reveal the ZBA feature which is discussed in this article.

3. Results and discussion

Before depositing the molecules, the pristine Al electrodes are characterized by recording opening and closing curves, i.e., measurements of the linear conductance as a function of the electrode distance, from which we calculate the conductance histogram. Typical traces and the resulting histogram obtained from the opening curves are shown in Fig. 1. In the middle panel five examples of opening traces are given, showing the typical sawtooth-like shape. The conductance of the last plateau corresponding to single-atom contacts ranges from 0.5 to 1.1 G_0 (where $G_0 = 2e^2/h$ is the conductance quantum) as indicated by the dotted black lines. These plateaus give rise to the first pronounced maximum around 0.8 G_0 with a natural width of $\pm 0.3\,G_0$. These findings are typical for clean Al junctions [32–37]. For the deposition of the molecules the electrodes are broken to form a tunnel contact with a gap of approximately 0.5 nm, which is the approximate diameter of a C_{60} molecule. After deposition of 1 ML of C_{60} the opening traces do still reveal a sawtooth behavior, but an additional step occurs below 0.5 G_0 (indicated by the dashed red line sin the lower panel), while the single-atom step around 0.8 G_0 remains visible, with somewhat shorter plateau length and slightly enhanced conductance, though. Not all opening curves display the additional step (see red and green curves in the lower panel). A sizeable fraction of opening traces shows multiple steps on the last plateau with conductance ranging from 0.4 to 0.8 G_0 as depicted by the magenta curve in the lower panel. As we will show below, these types of opening traces are typical for contacts showing a well-pronounced ZBA. As a result of these types of opening traces the red histogram shown in the top panel of Fig. 1 still reveals the multiple maxima at values slightly higher than found for clean Al. Additionally we find enhanced probability but no pronounced peak for $G < 0.5\,G_0$. The impact of the presence of C_{60} onto the histogram is much smaller than for C_{60} on Au, where the appearance of an additional peak in the histogram around $0.1\,G_0$ was found [31]. In similar experiments using a scanning tunneling microscope for contacting C_{60}, values ranging from 0.1 to $0.3\,G_0$ have been observed for the contact metals Au, Ag and Cu [14]. Even higher conductance values of C_{60} up to 3 G_0 when contacted with Al electrodes are expected because of the covalent character of the bond [30], but have not been observed experimentally yet. However, the absolute value strongly depends on the exact orientation of the electrode's crystal direction and configuration, and the orientation of the molecule in the gap. High conductance values correspond to strong coupling of the molecular orbitals to the metal electrodes, which in turn would enhance the probability to observe correlation effects like the Kondo effect, as long as the fullerene can still be charged. The Al–C_{60}–Al system is thus a good candidate for studying Kondo effect in the unconventional regime of intermediate to strong coupling, where the off-resonance conductance is in the range of 0.1 to 0.4 G_0.

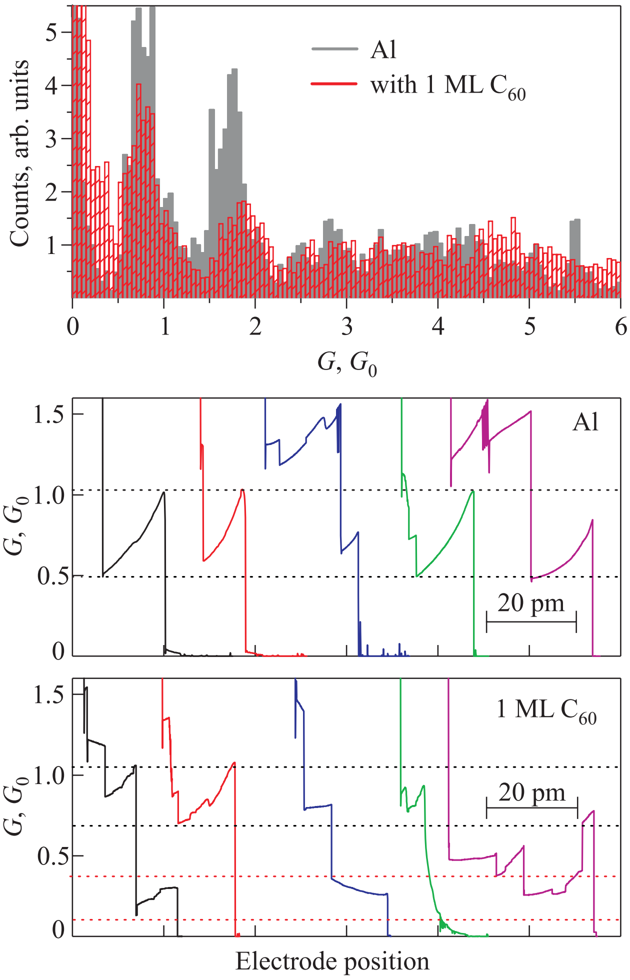

Fig. 1. (Color online) Top panel: Conductance histogram of an Al MCBJ (black) and of an Al MCBJ after deposition of 1 ML C_{60} (red). Both histograms were calculated from closing curves measured at 10 K in UHV. Central and bottom panel: Selection of opening traces before and after deposition of 1 ML C_{60}.

Before we describe our findings in the strong coupling regime, we note that in the intermediate coupling regime our findings are in line with the observations reported in Ref. 7. Figure 2 displays a set of dI/dV curves of a contact with an off-resonance conductance of $G_{or} = 0.028\,G_0$ for various temperatures. The resonance forms suddenly and the strengthens slightly upon further approaching the electrodes. Upon warming up at constant electrode distance, the amplitude of the ZBA diminishes while G_{or} slightly

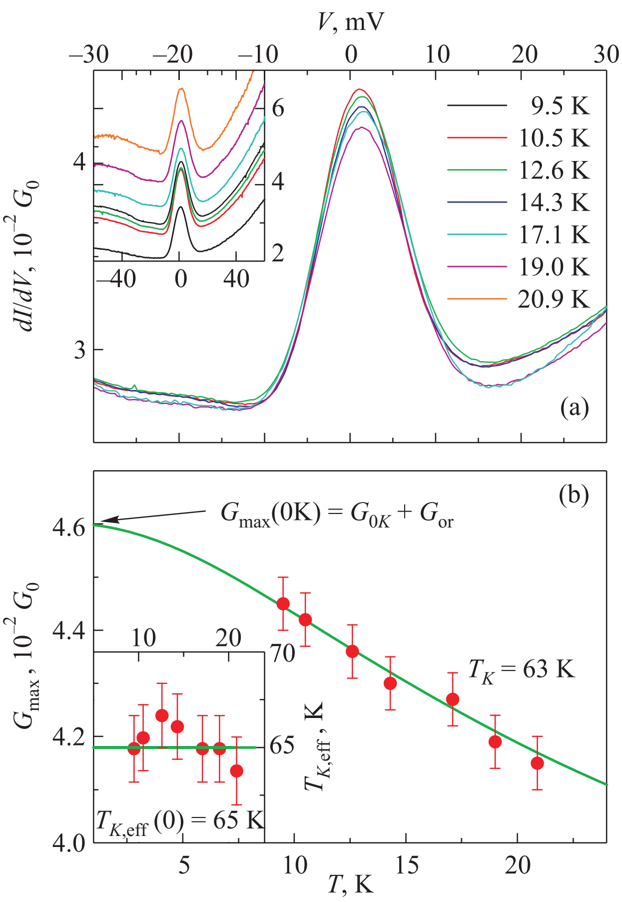

Fig. 2. (Color online) (a) Differential conductance curves of a Al MCBJ after deposition of 0.5 ML C_{60} at 10 K for different measuring temperatures and after subtracting the temperature dependence of the off-resonance conductance G_{or}. G_{or} at the lowest measurement temperature was $G_{or} = 0.028 G_0$. Inset: The same spectra at larger voltage scale showing the raw data before subtraction. (b) Peak conductance as a function of temperature and fit according to Eq. (3) yielding $T_K = (63 \pm 3)$ K. Inset: Effective Kondo temperatures $T_{K,\text{eff}}(T)$ determined from the FWHM of the spectra shown in the top panel and linear extrapolation to $T = 0$ resulting in $T_{K,\text{eff}}(0) = (65 \pm 3)$ K.

increases. For the further analysis we subtract this increase. We then follow the temperature dependence of the ZBA from our lowest effective measurement temperature until the contact gets lost due to thermal drift, diffusion, oxidation of the metal electrodes or other spontaneous rearrangements. This usually happens around 25 to 30 K. In the accessible temperature range the height of the ZBA, $G_{max}(T)$, decreases logarithmically with increasing temperature in agreement with the expectations for the Kondo effect as given in Eq. (3) [2]. We note, that despite this agreement, due to the limited temperature range this dependence is not sufficient to serve as an unambiguous proof of the Kondo effect. For the example shown in Fig. 2 we obtain $T_K = (63 \pm 3)$ K. The FWHM is mainly independent of the temperature in agreement with the expectation of the Kondo theory. Fitting a Lorentzian shape to the ZBAs results in a peak width FMHM $\simeq 10$ meV corresponding to $T_K = (65 \pm 3)$ K also in agreement with Kondo theory and previous findings for the Au–C_{60}–Au system [7]. Similar observations were made for all contacts with $G_{or} \lesssim 0.06\, G_0$. We now turn to our findings in the strong coupling regime. In Fig. 3(a) we show a set of dI/dV curves of a contact with G_{or} of $0.1\, G_0$ for various temperatures. A pronounced ZBA is observed, the amplitude $G_{max}(T)$ of which decreases with temperature. While the temperature dependence again follows the Kondo scaling function (3), the FMHW behaves differently. In this temperature range the FWHM depends linearly on the temperature as demonstrated in the inset. In the intermediate coupling regime the FWHM is expected to saturate at low temperatures, but exceptions of this expectation have been reported before [3,7]. Hence, for the strong coupling regime it is not clear, which temperature dependence should be expected. The linear extrapolation of the width FWHM(T) to $T = 0$ yields $T_{K,\text{eff}}(0) = (69.3 \pm 1.0)$ K in very good agreement with the result obtained from the fit to Eq. (3) that yields $T_K(0) = (69.0 \pm 1.0)$ K. However this

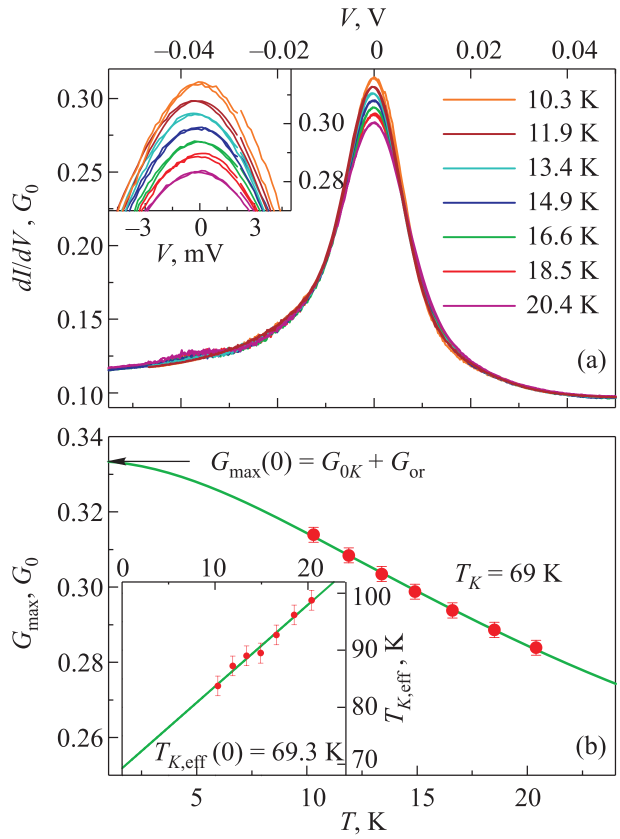

Fig. 3. (Color online) (a) Differential conductance curves of a Al MCBJ after deposition of 0.5 ML C_{60} at 10 K for different measuring temperatures showing a pronounced ZBA. Inset: Zoom onto the maximum. (b) Peak conductance as a function of temperature and fit according to Eq. (3) yielding $T_K = (69.0 \pm 1.0)$ K. Inset: Kondo temperatures $T_{K,\text{eff}}(T)$ determined from the FWHM of the spectra shown in the top panel and linear extrapolation to $T = 0$ resulting in $T_{K,\text{eff}}(0) = (69.3 \pm 1.0)$ K.

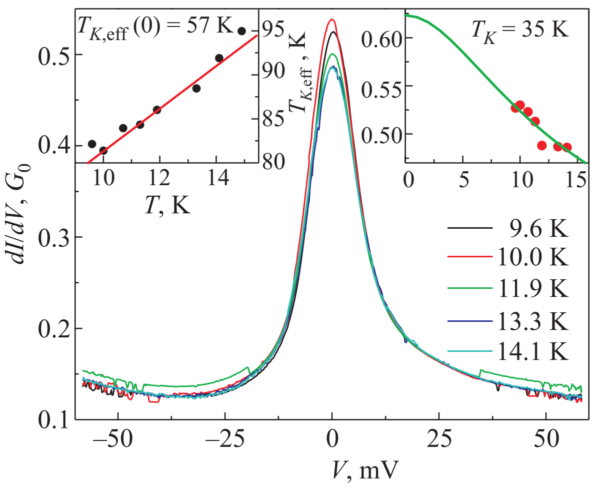

Fig. 4. (Color online) Differential conductance curves of an Al MCBJ after deposition of 0.5 ML C_{60} for different measuring temperatures showing a pronounced ZBA. Left inset: Effective Kondo temperatures $T_{K,\mathrm{eff}}(T)$ determined from the FWHM of the spectra shown in the main panel and linear extrapolation to $T = 0$ resulting in $T_{K,\mathrm{eff}}(0) = (57 \pm 2)$ K. Right inset: Peak conductance as a function of temperature and fit according to Eq. (1) yielding $T_K = (35 \pm 5)$ K.

agreement seems to be a mere coincidence as demonstrated with the next example, Fig. 4. Here the background conductance G_{or} is yet higher $G_{\mathrm{or}} = 0.13\, G_0$ and the extrapolation from the FWHM to zero temperature yields $T_{K,\mathrm{eff}} = (57 \pm 2)$ K while from the analysis of $G_{\max}(T)$ we obtain $T_K = (35 \pm 5)$ K.

For other Al–C_{60}–Al contacts we observe Kondo temperatures ranging from 35 to 160 K for off-resonance conductance values ranging from 0.007 to 0.4 G_0 with the clear tendency of higher T_K for higher G_{or}. The amplitudes $G_{\max}(0)$ range from 0.03 to 0.82 G_0. All examples follow the trend demonstrated by the three examples shown in Figs. 2, 3, and 4: For low $G_{\mathrm{or}} \lesssim 0.1\, G_0$ the expectations of the Kondo theory are fulfilled, while for higher $G_{\mathrm{or}} \gtrsim 0.1\, G_0$ the FWHM starts to be temperature dependent and higher than expected according to Kondo theory. No such ZBAs have been observed on contacts made from bare Al contacts. The noise floor of our experiment corresponds to 1% of the measurement range, i.e., resonances smaller than this value cannot be detected. Yet, since all kinds of ZBAs are observable on the same junction in the same cool-down run and even in the same closing curve, we argue that they have the same physical origin, namely Kondo screening of a spin-polarized state on the C_{60} molecule.

In order to test whether the observed ZBA is in fact caused by the Kondo effect we follow the development of the ZBA as a function of electrode distance for tuning the coupling strength Γ, Fig. 5. Changing the electrode distance influences the geometry of the junction on the atomic length scale and thus affects the wavefunctions of the conducting modes and therefore the charge transfer between the electrodes and the molecule. In the experiment shown in Fig. 5 the electrodes were closed in steps of 0.5 pm and dI/dV spectra were taken when the motion was stopped.

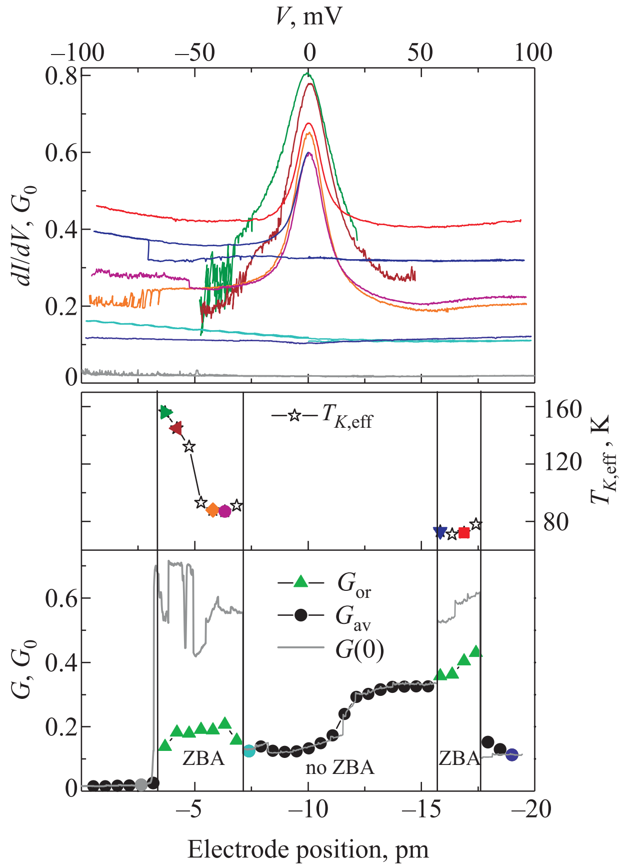

Fig. 5. (Color online) Development of the ZBA as a function of distance when closing an Al MCBJ from the tunnel regime, measured at 9.3 K. Top panel: Selected spectra measured at distances marked in the lower panels with a symbol in the respective color. Central panel: Effective Kondo temperature $T_{K,\mathrm{eff}}$ deduced from the FWHM. Bottom: Corresponding closing trace: Symbols: off-resonance conductance G_{or} for those spectra with ZBA, and average conductance G_{av} for those junctions without ZBA; grey line: conductance measured at 0 mV.

The measurement was performed at $T = 9.3$ K. A selection of spectra which reveal the ZBA as well as a selection of those without are shown in the top panel of Fig. 5. The bottom panel displays the closing curve on which the spectra have been observed. For the spectra without ZBA we plot the average conductance G_{av} as closed circles, while closed triangles represent the values of the off-resonance conductance G_{or}. In addition we show the trace of the maximum conductance $G(0)$ measured around 0 mV as grey line. As expected for the regions without ZBA G_{av} and $G(0)$ are in very good agreement, while in the regions with ZBA $G(0)$ exceeds G_{or} markedly and shows considerably more steps, comparable to the last step in the magenta curve in the lower panel of 0. For the spectra with ZBA we determine $T_{K,\text{eff}}$ from the FWHM(T) as described above and plot it as open stars in the central panel. The color code of the symbols corresponds to the one of the traces in the top panel. The spectra have been measured in the following manner: The voltage is swept from 0 V to +100 mV, then to –100 mV and finally back to 0 V. For clarity, for most spectra we only show the central part of the sweep. We set the electrode distance to 0, where the conductance exceeds a value of 0.01 G_0 and count. Up to electrode distance 0 to –3.4 pm and for a conductance of up to 0.02 G_0 no ZBA is observed (grey spectrum in top panel). At electrode position –3.5 pm, when the G_{or} has increased to 0.13 G_0 a ZBA appears spontaneously (dark green curve) with very high resonance amplitude $G_{\max} = 0.8\,G_0$ and relatively large width corresponding to $T_{K,\text{eff}} = 157$ K. Above $|V| \approx 40$ mV the spectrum is rather noisy, presumably due to current-driven instabilities. Upon further approaching the electrodes, the ZBA remains present but its width and amplitude shrink and the noise disappears (brown, orange and magenta curve) until it suddenly disappears around $d = -7$ pm (light blue curve). The conductance is slightly smaller and increases again when continuing closing. At a distance of –15.5 pm and $G_{\text{or}} = 0.35\,G_0$ the ZBA reappears (blue curve) and vanishes again at $d = -17.5$ pm (purple curve). When continuing closing the contact, no further ZBA was observed.

The most important observation is that the ZBA vanishes and reappears suddenly upon very small changes of the electrode distance. These changes may occur during the measurement of a spectrum (without intentional change of the electrode distance, as, e.g., exemplified by the blue curve) and faster than the time required to take one data point in the dI/dV (50 ms). In Ref. 7, a gradual influence of the electrode distance onto T_K seemed to be the generic case, although a sudden jumping-in and out of the ZBA was also reported but not discussed. The continuous change of the ZBA parameters with electrode distance was described as changes of the coupling strength Γ with the distance and was more pronounced than in our experiment. The stability of our setup is sufficient to exclude drifts exceeding displacements of 0.1 pm, corresponding to only a small fraction of an atomic diameter and the typical extension of electronic wave functions. We therefore exclude the sudden changes of the ZBA to be caused by sudden changes of Γ. We interpret the sudden changes of the ZBA to be caused by charging and discharging of the molecule, which is possible on very short time scales. The spontaneous charging/discharging is presumably caused by fluctuations of the background charges, that in turn change the potential of the molecules such that the charged/uncharged state becomes the energetically most favorable. These background charge fluctuations are a well-known phenomenon causing instabilities that hampered applications of single-electron transistors in metrology [38]. This is supported by the fact that these sudden appearances/disappearances become less frequent when the sample is kept cold for longer time spans, allowing the two-level fluctuators two condense to their ground state. A third indication for this interpretation is that the values for T_K in both regions are similar (after settling and stabilizing after formation of the molecular contact), meaning that Γ and ε_0 are mainly unchanged. In the example displayed in Fig. 5, T_K ranges from ≈ 71 to 92 K which can be accounted for by changing the coupling strength by only 6% when assuming that the same electronic level of the dot is responsible for the Kondo effect. A slight variation of the coupling in this order of magnitude is very likely to occur when varying the distance and is even smaller than required to account for the changes of G_{or} and G_{av} over the same distance.

Another possible explanation of repeated appearance and disappearance of the ZBA would be acquiring higher charge states. In that case the first region with ZBA might correspond to one extra electron on the dot with spin 1/2, the subsequent region without ZBA would correspond to two electrons on the dot coupling to either spin 0 or spin 1. In both cases there would be no observable Kondo effect because the expected Kondo temperatures would be well below our measuring temperature. The second region with ZBA could consequently be a molecule charged with three electrons with total spin of 1/2 or 3/2, respectively. In both cases the expected Kondo temperatures are in agreement with our observation. However, it appears very unlikely that the molecule would be doubly or triply charged because the corresponding charging energy amounts to half an eV or more, that is very unlikely to be provided by background charge fluctuations or other spontaneous effects.

Finally, we note that we cannot completely exclude that the ZBAs observed at higher or have no magnetic origin at all, despite the fact that similar observations, i.e., ZBAs with temperature dependent FWHM have been interpreted as such [7]. Unfortunately the required magnetic fields necessary for observing a sizeable splitting of the Kondo resonance are much to high to be achievable experimentally for the rather elevated Kondo temperatures found here. However this is a very unlikely scenario, since the ZBAs at low or fulfill the Kondo theory predictions and occur on the same sample and same closing curve.

An alternative explanation of the high-conductance ZBA could be the formation of a resonant conductance state of the C_{60} with the conduction electrons of the Al electrodes [39], although it is then not straightforward to understand the spontaneous appearance and disappearance in that case.

Also this observation would be novel and promising, since in general molecules have a pronounced HOMO-LUMO gap and do not show perfect level alignment with the Fermi seas of the electrodes. The system Al–C_{60}–Al would then be a candidate for a highly conducting molecular device. Since the zero-bias conductance with and without the resonance differs markedly and since the width of the resonances is sufficiently large, this system might be used as a charge detector. Another conceivable application would be a conductance switch in the case that the charge state of the molecule can be controlled by external means like a gate electrode.

4. Summary

In conclusion, we reported the observation of preferred conductance values of Al–C_{60}–Al single molecule junctions of up to 0.4 of the quantum of conductance measured around 10 K. Superimposed on this conductance we observe pronounced zero bias anomalies. For rather low background conductance $G \lesssim 0.07\, G_0$ the shape and the temperature dependence of these resonances are in accordance with the expectations for spin 1/2 Kondo resonances with Kondo temperatures in the range of 35 to roughly 60 K. For higher conductance the shape of the width of the zero-bias anomaly becomes linearly dependent on the measurement temperature. The amplitude of the resonances may adopt values similar to the conductance of single-atom Al contact and can be slightly tuned by gentle changes of the electrode distance. However, occasionally we observe abrupt disappearance and reappearance of the resonances upon very tiny changes of the electrode distance. This finding gives support to the interpretation that the resonances are in fact caused by the Kondo effect involving an extra electron on the C_{60}, because both discharging or addition of an electron would completely quench the Kondo peak. However, the fact that for rather well-coupled contacts the resonance width decreases linearly with the temperature is not in agreement with the theory for the intermediate coupling regime. We suggest that the theories would have to be extended to cover the parameter range of our experiment.

Acknowledgments

We thank J.J. Palacios and N. Andrei for helpful discussions. Financial support by the Deutsche Forschungsgemeinschaft through SFB513, SFB767, SPP1243 and the Krupp foundation through the Alfried-Krupp-Förderpreis für Junge Hochschullehrer is gratefully acknowledged.

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