First publ. in: Physical Review Letters 99 (2007), 086801

Influence of Laser Light on Electronic Transport through Atomic-Size Contacts

D. C. Guhr, D. Rettinger, J. Boneberg, A. Erbe, P. Leiderer, and E. Scheer*

Fachbereich Physik, Universität Konstanz, D-78457 Konstanz, Germany

(Received 23 January 2007; published 21 August 2007)

This Letter reports on the influence of laser irradiation onto the electrical conductance of gold nanocontacts established with the mechanically controllable break-junction technique. We concentrate on the study of reversible conductance changes which can be as high as 200%. We investigate the dependence on the initial conductance of the contacts, and on the wavelength, the intensity, and the position of the laser spot with respect to the sample. Under most conditions an enhancement of the conductance is observed. Several physical mechanisms which might contribute to the observed effect including thermal expansion, rectification, plasmon excitation, and photon-assisted transport are discussed, among which the two latter ones are most likely the dominating ones.

Metallic nanostructures with fine tips and sharp edges are interesting candidates for optical antennae due to possible field enhancement (FE) effects at the parts with small curvature radii and narrow gaps [1]. The excitation of resonant plasmons can amplify the electrical field at positions within or close to the nanostructures. The field enhancement in bow-tie shaped metal structures [1] has been calculated and detected by several near field optical methods [2] and direct imaging via ablation of the underlying substrate [3]. Here we address the question whether FE can be detected in the electronic transport between two metallic tips connected via an atomic-size contact. For this geometry to our knowledge no calculations of the magnitude of the FE do exist; however, it can be assumed that as long as the coupling between the two electrodes is weak, it should still exist. Such contacts can be realized via mechanically controllable break junctions (MCBs), i.e., free-standing nanobridges with lateral sizes in the order of 100 nm and bridge lengths of several micrometers. By bending the substrate the bridge is elongated and its cross section is reduced until it finally breaks. Just before rupture, the constriction forms a contact with one or a few atoms in cross section. These contacts represent model systems which are regularly used for revealing and understanding the electronic transport properties of atomic-scale circuits [4].

Besides this aspect, MCBs can be broken to form electrode pairs with a narrow gap in the order of picometers to nanometers between two very fine metallic tips. Such electrodes are used for contacting individual nano-objects such as quantum dots, clusters, or molecules [5]. The knowledge of the behavior of the electrodes is a mandatory prerequisite for the interpretation of molecular-junction transport experiments under laser irradiation [6].

In our work we therefore concentrate on the study of laser illumination onto the metal electrodes alone. Already for this conceptually rather simple device the influence of the laser light may be manifold. One rather trivial effect is the geometry change of the tips due to thermal expansion because of the deposited energy of the laser pulse. This effect has been shown to be the dominating one for a scanning tunneling microscope (STM) under pulsed laser-light irradiation [7,8]. However, for the MCB geometry this effect is expected to be much smaller due to the very small dimensions of the free-standing bridge arms.

The electromagnetic wave of the laser pulse represents a high-frequency electrical field that is coupled to the metal bridge. The ac field may create an asymmetric contribution to the dc current for those voltages where the I-Vs are nonlinear [9]. Asymmetric irradiation of the contact might create temperature gradients—and thus thermocurrents—or photocurrents. Last but not least, photon-assisted transport (PAT), i.e., the creation of quasiparticles with an energy $\hbar\omega$ above the Fermi energy, will create a nonequilibrium in the electronic system. This means that high-energetic quasiparticles may contribute to the transport. Because of the energy dependence of the transmission coefficients of the conduction channels [10], the resistance of the contact will change [11].

The samples are fabricated by electron-beam lithography along the lines of Ref. [12]. The suspended nanobridges are 2 μm long and 100 nm thick made of Au with a 200 nm $\times$ 100 nm constriction in the center. We use an argon-krypton cw laser as light source, which allows us to select one of several wavelengths in the range between 480 nm and 650 nm. The sample is mounted on a breaking mechanism inside a UHV chamber at room temperature with a base pressure of 10^{-9} mbar in order to avoid contamination of the junction. The mechanics are designed such that the position of the nanobridge remains constant upon bending the substrate in order to ensure stable optical focal conditions. The UHV chamber is equipped with a fused silica window for optical access. If not stated differently, the polarization of the laser light was chosen parallel to the current for the measurements presented here. For perpendicular polarization similar results but with an amplitude that is smaller by about a factor 2 to 3 are observed. A more detailed investigation is necessary in order to quantify the exact influence of the polarization. The sample is connected with a 100 kΩ series resistor to a

dc-voltage source in order to perform a current bias when the sample is in the few-atom regime (several kΩ). We measure the signal across the sample in dc as well as in ac mode. The amplitude of the ac part, caused by each light pulse, is measured by a sample-and-hold circuit (S&H) with an integrating amplifier.

Figure 1 shows a current-voltage (I-V) characteristic of a contact for a conductance without irradiation of $G_i = 11G_0$, where G_0 denotes the conductance quantum $G_0 = 2e^2/h$. At several current values laser pulses with duration $\tau = 700$ μs, $\lambda = 488$ nm, and power $P = 0.85$ mW are shed onto the contact. During the pulse the absolute value of the voltage measured across the contact is reduced by the same amount for both polarities. The amplitude of the laser induced voltage drop is proportional to the current. Neither considerable current nor voltage offsets nor asymmetries are observed, indicating that the effect of the light is a bare change of conductance to $G_f = 17G_0$. The fact that the conductance change is observed for linear I-V characteristics indicates that it is not created by a rectification effect as observed in Ref. [9]. Pulse lengths and intensity are chosen such that the conductance change is completely reversible.

However, continuous irradiation of the device with $\lambda = 488$ nm for several seconds with a similar power ($\approx$ mW) results in irreversible conductance changes (not shown), indicating that atomic rearrangements are triggered by the cw light.

When elongating the bridge without irradiation, its conductance G_i decreases in steps of the order G_0, their exact sequence changing from opening to opening because the atomic arrangement of the central region differs for each opening process [4]. The preferred conductance value of single-atom gold contacts is $1G_0$ [4].

The light-induced signal (LIS) varies from contact to contact. In order to deduce the typical behavior, we simultaneously measure the conductance and the LIS as a relative conductance change $\Delta G/G_i = (G_f - G_i)/G_i$ upon continuously opening the bridge under pulsed-light irradiation with a repetition rate of 50 Hz. Figure 2 gives an example of such measurements recorded for green light with $\lambda = 515$ nm. As usual for room-temperature experiments on lithographic MCBs, the conductance plateaus are not always very well pronounced. The tunneling regime is difficult to access when opening because the contact tip atoms relax back upon breaking giving rise to strong decrease in the conductance by several orders of magnitude [4].

The LIS is positive throughout the whole opening process. It reaches a maximum with large fluctuations for $G \simeq 2G_0$. The inset of Fig. 2 shows a zoom of the final part of the opening trace. Obviously the local maxima of the LIS correlate with the local maxima of G while the overall development is roughly anticorrelated: the LIS increases for decreasing G_i up to its maximum at a few G_0. For smaller G_is it decreases again and becomes very small or slightly negative in the tunneling regime.

A priori it is not predictable whether in our geometry thermal expansion should give rise to an enhancement or a reduction of the conductance, since the laser spot also hits the underlying substrate. If the thermal expansion of the nanobridge is larger than the one of the substrate, the two electrodes are pushed together; if the thermal expansion of the substrate exceeds the one of the nanobridge, then the two tips are pulled apart from each other. Whether the motion of the tips results in an enhancement or reduction of G_i depends on the plateau shape of the conductance trace upon opening: for a horizontal plateau the conductance is constant upon stretching the contact; i.e., changing the bridge length does not give any conductance change. For negatively declined plateaus, i.e., decreasing G_i upon stretching, an elongation of the tips yields an enhancement of G_i while positively declined plateaus give rise to a decreasing G_i when pushing the tips together. The con-

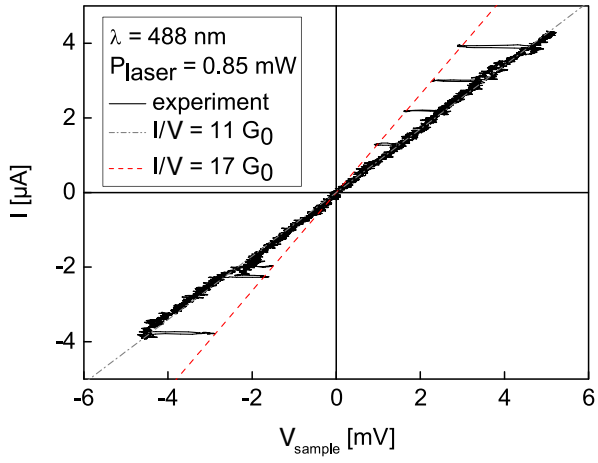

FIG. 1 (color online). Current-voltage (I-V) characteristic of a contact with $G_i \approx 11G_0$ showing the influence of several light pulses with $P = 0.85$ mW and $\lambda = 488$ nm. During the light pulses the conductance is enhanced as indicated by the guide to the eye showing a linear I-V characteristic for $G_f \approx 17G_0$.

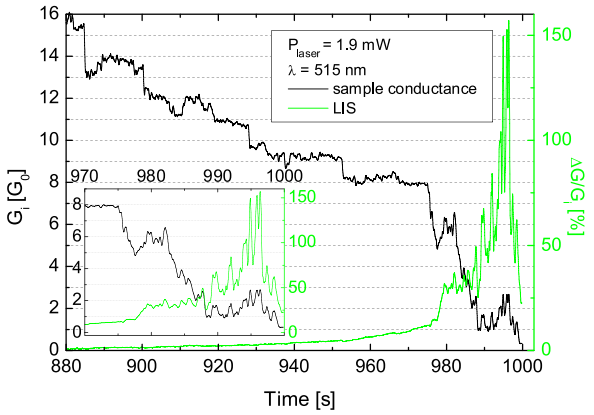

FIG. 2 (color online). Conductance and light-induced relative conductance change $\Delta G/G_i$ vs time when opening the break-junction continuously. Inset: close-up of the few-atom region with $G_i < 9G_0$.

ductance trace of Fig. 2 shows horizontal as well as negatively and (only rarely) positively declined plateaus. Nevertheless, in our experiments the LIS is almost always found to be positive (see below). Furthermore, the strong exponential distance dependence of G_i for vacuum tunneling should give rise to a large relative conductance change upon thermal expansion due to the irradiation. However, the LIS is usually smallest in this regime (not shown). Furthermore, we show below that, when the laser beam is focused to the side of the bridge, a small negative LIS can be observed. Hence, although thermal expansion is very likely to be present in our experiment, we conclude from these considerations that it gives only a minor contribution to the observed LIS because it does not explain an always positive LIS or a larger signal in the contact regime as compared to the tunneling situation.

Figure 3 depicts the averaged LIS (over 170 opening traces each) recorded on the same sample with the same laser power $P = 2.2$ mW for four light wavelengths. For each opening trace the LIS is related to the initial conductance value G_i. The averaged LIS and its standard deviation are then plotted on a logarithmic scale versus G_i. As seen from Fig. 3 the signal is largest for blue light with $\lambda = 488$ nm and decreases with increasing wavelength. The LIS also depends on the conductance: it is largest for few-atom contacts and decreases for smaller and larger contacts. The G_i value for which the maximum signal is observed does not depend systematically on λ. In particular, for $\lambda = 488$ nm the standard deviation is markedly larger for $G_i \gtrsim 5G_0$ than for $G_i \lesssim 5G_0$ although the amplitudes are similar. This indicates that the size of the LIS depends on the detailed atomic arrangement. For larger G_i the possibilities to form a contact are manifold, while contacts with small G_i have only a few atoms in cross section.

The amplitude dependence of the LIS on λ roughly correlates with the variation of the reflectivity of gold with λ, which increases from 39% for $\lambda = 488$ nm up to 97% for $\lambda = 648$ nm. This means that the deposited energy decreases by a factor of ≈ 20, while the maximum signal size decreases by about a factor of 40.

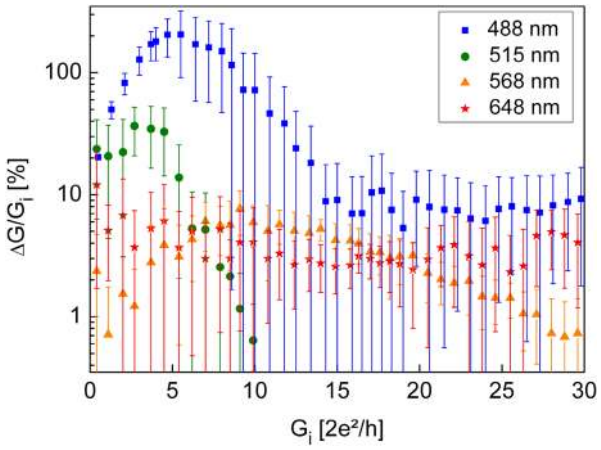

FIG. 3 (color online). Averaged light-induced signal $\Delta G/G_i$ vs G_i for different laser wavelengths λ and $P = 2.2$ mW.

Further information about the effect is obtained from recording a map of the signal size with respect to the position of the laser spot. The maps for $\lambda = 488$ nm and 648 nm (averaged over several contacts with a conductance of $G_i = 6.5G_0$) and polarization perpendicular to the transport direction are shown in Fig. 4. A clear position dependence is observable for all wavelengths (data for $\lambda = 515$ and 568 nm not shown). In Fig. 4 we superimposed a drawing of the sample geometry with the recorded LIS. For $\lambda = 488$ nm the largest signal is observed when the center of the laser spot is located on the leads within a distance of 10 μm from the contact. It is roughly symmetrical with respect to the contact. For $\lambda = 648$ nm a relative minimum is observed right at the position of the contact, and the largest signal is observed when the spot is located 20 μm away from the bridge, where the leads become wider. More than a diameter of the laser spot aside of the contact a small negative LIS is observed, indicating that the thermal expansion of the substrate gives rise to a *negative* LIS in the order of only a few percent of the total signal. Thus, the observed enhancement when the laser spot does hit the metallic bridge is due to light coupled into the metallic bridge. When irradiating the narrow constriction, only a small part of the laser energy is coupled into the metallic bridge, but the majority is absorbed by the substrate. Thus, the minimum in the LIS can be explained by the geometry of the experiment. Contrary, the lack of a minimum for the blue light indicates that an energy dependent mechanism is at the origin of the strong positive LIS.

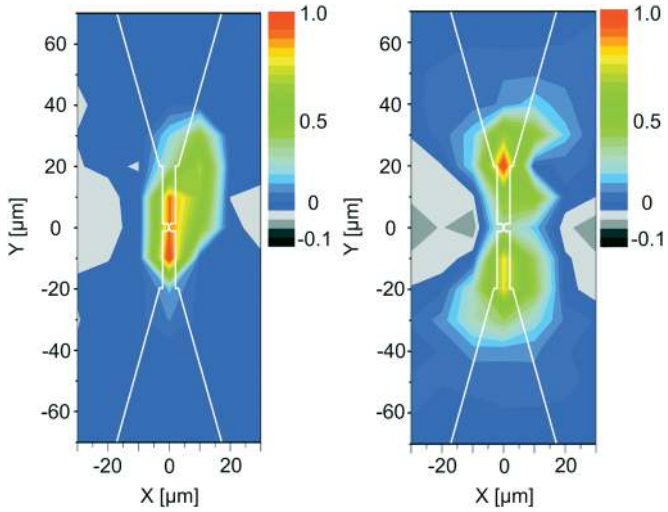

FIG. 4 (color). Position map of $\Delta G/G_i$ for left (right) $\lambda = 488$ nm (648 nm) for $G_i = 6.5G_0$, $P = 2.2$ mW (23 mW), and polarization perpendicular to the current. The signal has been determined by averaging over 20 (10) consecutive light pulses for each position. The color scale is normalized to the maximal signal which was 64% (94%) for 488 nm (648 nm). As a guide to the eye we show superimposed the contour of the sample (white lines).

A possible explanation could be PAT reflecting the energy dependence of the transmission coefficients of the conduction channels as proposed by Viljas and Cuevas [11]. The mechanism is such that the incoming photons are absorbed by individual electrons, creating quasiparticles (QPs) with energy $\hbar\omega$ below and above the respective Fermi energies E_F of both electrodes. Since electrons on both sides of the constriction are concerned, the QPs may travel in both directions. In the theory the effect is treated as an additional ac voltage at frequency ω superimposed to the transport voltage, but no further details of the light field like, e.g., its polarization, are taken into account. The same approach has been used before successfully to explain the influence of microwaves onto the transport properties of atomic-size contacts [13]. In this low-energy regime the influence of the microwave irradiation is only detectable by the study of superconducting properties, a phenomenon which involves typical energies in the order of meV. Since the photon energy in the visible regime corresponds to the typical energies of electronic band structures in metals, irradiation with these energies enables to directly probe the local density of states. The transmission coefficients of the conduction channels reflect the local density of states at the central atom. Although the exact functional dependence of the transmission differs from that of the density of states, the typical energy scales are identical. For single-atom contacts of Au it has been predicted that the typical energy scale for non-negligible variation of the transmission coefficients is about one to several electron volts [14]. Those QPs have a lifetime in the order of several ten femtoseconds, depending on their energy [15]. It has been shown that the effective thermalization times are much longer than the lifetimes estimated from Fermi liquid theory [15–17]. Since furthermore the transport mode and the velocity of the highly excited QPs differs from the thermal ones close to the Fermi edge, it is difficult to determine a corresponding range or scattering length [15]. A rather optimistic estimate yields a mean free path in the order of a few hundred nanometers. This means that mainly incoming photons within this distance from the contact will contribute to the LIS. For photon energies of a few eV, variations of the conductance are thus expected, the sign of which depends on the particular energy landscape of gold atomic-size contacts [11]. The dominating transmission of the s band decreases for smaller as well as for higher energies, but for lower energies the $5d$ bands and for higher energies the $6p$ bands start to contribute to the transport. The order of magnitude of our experimental LIS is in accord with the findings of Ref. [11]. However, a quantitative analysis in terms of PAT is not possible yet because of the simplified assumptions of the model calculation and the lack of information about the detailed atomic structure in the experiments.

Because of the short mean free paths of the high-energy QPs one would thus expect to have a maximal signal closest to the contact, but because of the reduced lateral size less photons arrive at the metal bridge. For $\lambda = 488$ nm the increase of the efficiency onto the transport and the loss in number of photons seem to compensate. The photon energy for $\lambda = 648$ nm is below the plasma edge $\hbar\omega_p \simeq 2.5$ eV (corresponding to $\lambda \approx 500$ nm) [18], while the one for $\lambda = 488$ nm is above. Thus, for $\lambda = 648$ nm the incident light wave may excite plasmons which have a typical range in the order of tens of micrometers, while above $\hbar\omega_p$ the photon energy is preferentially transferred to individual electrons. A possible explanation of the strong LIS when the focus is located 20 μm away from the bridge for $\lambda = 648$ nm is thus the redistribution of the occupation of the electronic states due to the creation of plasmons. Further experiments are necessary to verify this interpretation.

In conclusion, we have presented electric transport measurements carried out on atomic-size contacts connecting the two parts of a bow-tie shaped optical-antenna geometry under irradiation with laser light. We observe strong, contact specific conductance changes, the amplitude of which strongly depends on the wavelength of the laser. The observations can be understood by a recently predicted photon-assisted transport mechanism and the creation of plasmons and are important for the interpretation of light-induced effects in molecular-electronics devices.

Fruitful discussions with W. Belzig, J. C. Cuevas, J. K. Viljas, and A. Leitenstorfer are gratefully acknowledged. This work was financially supported by the Deutsche Forschungsgemeinschaft through SFB 513 and the Alfried Krupp von Bohlen und Halbach Foundation.

*Corresponding author.
elke.scheer@uni-konstanz.de

[1] J. N. Faharani *et al.*, Phys. Rev. Lett. **95**, 017402 (2005).
[2] P. Mühlschlegel *et al.*, Science **308**, 1607 (2005).
[3] P. Leiderer *et al.*, Appl. Phys. Lett. **85**, 5370 (2004).
[4] N. Agraït *et al.*, Phys. Rep. **377**, 81 (2003).
[5] *Introducing Molecular Electronics*, edited by G. Cuniberti *et al.* (Springer, New York, 2005).
[6] D. Dulić *et al.*, Phys. Rev. Lett. **91**, 207402 (2003).
[7] S. Grafström *et al.*, J. Appl. Phys. **83**, 3453 (1998).
[8] J. Boneberg *et al.*, Appl. Phys. A **66**, 615 (1998); **67**, 381 (1998).
[9] R. Möller *et al.*, J. Vac. Sci. Technol. B **9**, 506 (1991).
[10] R. Landauer, Philos. Mag. **21**, 863 (1970).
[11] J. K. Viljas and J. C. Cuevas, Phys. Rev. B **75**, 075406 (2007).
[12] J. M. van Ruitenbeek *et al.*, Rev. Sci. Instrum. **67**, 108 (1996).
[13] M. Chauvin *et al.*, Phys. Rev. Lett. **97**, 067006 (2006).
[14] J. C. Cuevas *et al.*, Phys. Rev. Lett. **81**, 2990 (1998).
[15] J. Cao *et al.*, Phys. Rev. B **58**, 10 948 (1998).
[16] M. Aeschlimann *et al.*, Appl. Phys. A **71**, 485 (2000).
[17] C. Guo *et al.*, Phys. Rev. Lett. **86**, 1638 (2001).
[18] J. H. Simmons and K. S. Potter, *Optical Materials* (Academic, New York, 2000), p. 75.