Eur. Phys. J. Appl. Phys. **31**, 191–194 (2005)
DOI: 10.1051/epjap:2005045

THE EUROPEAN PHYSICAL JOURNAL APPLIED PHYSICS

Calorimetric tunneling study of heat generation in metal-vacuum-metal tunnel junctions

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Received: 30 November 2004 / Accepted: 15 April 2005
Published online: 18 August 2005 – © EDP Sciences

Abstract. We propose a novel calorimetric tunneling (CT) experiment allowing exact determination of heat generation (or heat sinking) in individual tunnel junction (TJ) electrodes, which opens new possibilities in the field of design and development of experimental techniques. Using such an experiment we have studied the process of heat generation in normal-metal electrodes of the vacuum-barrier tunnel junction (VBTJ). The results show there exists a dependence of the mutual redistribution of the heat on applied bias voltage and the direction of tunnel current, although the total heat generated in the tunnel process is equal to the Joule heat, as expected. Moreover, the present study indicates generated heat represents the energy of non-equilibrium quasiparticles coming from inelastic electron processes accompanying the process of elastic tunneling.

PACS. 73.40.Gk Tunneling – 73.40.Rw Metal-insulator-metal structures – 72.15.Jf Thermoelectric and thermomagnetic effects – 07.79.-v Scanning probe microscopes and components

A deeper understanding of the power dissipation due to charge injection in tunnel junctions (TJs) and point contacts (PCs) is of great importance for optimal design, fabrication and further miniaturization of nano-scale electronic devices. One of the important points is the detailed knowledge of the nature of the heat generation and an appropriate characterization of energy dissipation processes in such structures. Experiments with ballistic PCs [1] show asymmetry of heat generation associated with the fact that the charge carriers accelerated in the field region of the contact propagate ballistically, i.e. without scattering, and dissipate gained energy generating non-equilibrium quasiparticles only *after* passing the contact [2,3]. This asymmetry can even be a source of information of electron-phonon interaction [3,4]. Observations of dissipation asymmetries in TJs at high bias [5] and studies of self-heating due to electron tunneling in the Coulomb-blockade electrometer [6], as well as investigations of electronic refrigeration in the normal-metal – insulator – superconductor (NIS) TJ [7] show that the process of charge tunneling is frequently associated with generation of thermal gradients. This shows the need for experimental studies yielding information about heat generation or heat sinking in individual TJ electrodes.

Analyzing properties of a tip-sample configuration, as well-known from scanning tunneling microscopy (STM), we are of the opinion that utilization of the VBTJ should allow an exact calorimetric determination (measured in absolute units) of the heat power dissipated in each of the TJ electrodes as the vacuum tunnel barrier secures a thermal decoupling of the electrodes. Moreover, unlike experiments where the TJ is placed e.g. in liquid helium [5], the studies performed at vacuum conditions remove problems coming from parasitic thermal coupling of the TJ electrodes with their surroundings.

In this paper we explain the principle of the proposed CT experiment and present results of exact calorimetric measurements of the heat generation in the *separate* electrodes of normal-metal – vacuum – normal-metal TJ obtained by this novel experimental technique. Our results show that the mutual redistribution of the heat power dissipated in (normal) metal electrodes of a TJ depends on the bias voltage applied over the TJ and it can be adequately described within the generally accepted conception of electron tunneling [5,6,8].

In principle, the CT experimental setup consists of the STM-like tunneling unit and a sample holding calorimeter for taking specific heat measurements by the relaxation method, e.g. as used in [9,10]. The experimental setup used for the studies presented in this paper is shown in Figure 1. The tip is connected to the linear piezo-positioner [11] of the low temperature high vacuum compatible tunneling head. Its separation from the sample fixed to the sapphire plate is controlled by the z-feedback STM control electronics [11]. The sapphire plate is equipped with a bare chip Ge-thermometer and a RuO_2 resistor as a heater, supported from one side by three stainless steel needles fixed to the copper block.

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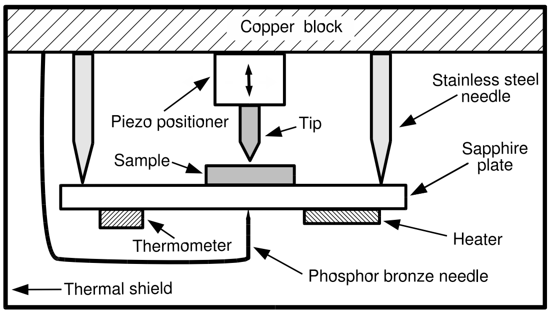

Fig. 1. Schematic depiction of the CT experimental setup.

From the other site it is pressed by a warped phosphor bronze needle thermally linked to the same copper block. The sample, thermometer and heater are glued to the plate using a small amount of GE-7031 varnish. Such a design, similar to a calorimeter used for specific heat studies by the relaxation method [10] and calorimetric absorption spectroscopy studies [12] has shown sufficient mechanical stability and plausible heat resistance for CT experiments at temperatures up to 6 K at least.

Both the tip and the sample were cut from the same gold based alloy $Au_{0.7}Cu_{0.16}Ag_{0.14}$ in order to prevent formation of an insulating layer on the tip and the sample surfaces and to avoid effects due to dissimilarities of TJ electrodes. Immediately after cleaning of the tip and the sample surfaces the experiment was placed into the vacuum space of a ^{4}He cryostat, pumped to a high vacuum pressure with simultaneous overheating to a temperature above 40 °C and then slowly cooled to low temperatures.

The data were taken at temperatures close to 5.3 K. The heat power generated in the sample due to the tunnel current I was derived from the increase of the sapphire plate temperature. The experimental procedure consisted of the determination of the generated heat power P_{pos} and P_{neg} for positively and negatively biased sample, respectively, at the same absolute value of tunnel current I, with the aim to get an *exact comparison* of the heat power generated in the sample due to electron tunneling for "direct" and "reverse" polarity of the tunnel current.

Measurements at constant absolute value of I (Fig. 2) show that in the low-voltage limit ($V < 300$ mV) P_{pos} and P_{neg} are equal within the resolution of the experiment. At higher bias voltage V the heat power generated in the sample shows a clear asymmetry with respect to the orientation of I. The data show that P_{pos} (electrons injected *into the sample*) is greater than P_{neg} (electrons injected *from the sample*). The difference between P_{pos} and P_{neg} *nonlinearly* increases with increasing V. On the other hand, P-I dependencies at constant absolute value of V (inset of Fig. 2) show that the ratio P_{pos}/P_{neg} does not depend on I, as both P_{pos} and P_{neg} are linear functions of I within the resolution of experiment and scanned range of parameters. Because of the same material of the tip and the sample, no effects due to dissimilarities of the TJ electrodes are considered and the total power generated in the VBTJ can be expressed as $P_{tot} = P_{pos} + P_{neg}$. As shown in Figure 2, including its inset, there is an excellent

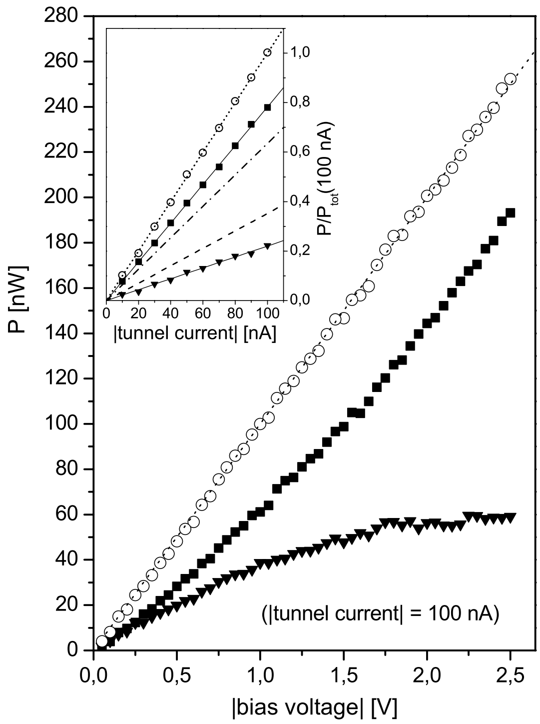

Fig. 2. Dependence of the heat power on the bias voltage for positively (squares), negatively (triangles) biased sample, and of the total power $P_{tot} = P_{pos} + P_{neg}$ (circles). The dotted line represents the power obtained using the classical formula $P_{Joule} = VI$. Inset: Dependence of normalized heat power $P/P_{tot}(100$ nA) on the tunnel current for positively (squares) and negatively (triangles) biased sample measured at $V = \pm 2.5$ V. Normalized total power (circles) is compared with that calculated using the formula $VI/(V \times 100$ nA) (dotted line). For illustration, normalized linear fits (by means of regression formula $y = bx$) of data taken at $V = \pm 1.0$ V for positively (dot-dashed line) and negatively (dashed line) biased samples are plotted as well.

coincidence of P_{tot} with Joule heat power $P_{Joule} = VI$ calculated for an electrically equivalent resistor replacing TJ. Here it should be noted that the estimation of Joule heat in the sample and in the tip due to their resistances, which are less than 1 Ω, yields a value less than 10^{-14} W. Taking into account that the estimated resolution of our experiment is ≈5 nW, the observed effects cannot be associated with Joule heat generation due to non-zero resistances of TJ electrodes.

The results represent a direct calorimetric verification of the model of energy dissipation in TJ as presented in [5] that can be summarized as follows. Considering metal P and metal N as the positive and negative TJ electrode, respectively, separated by a vacuum tunnel barrier as shown in Figure 3, the net tunnel current I can be expressed in the form

$$I = e \int_{-\infty}^{+\infty} n(E) dE. \quad (1)$$

Here $n(E)$ is the resulting number of tunneled electrons per one second within the energy range $\langle E, E + dE\rangle$ (see Ref. [13]) where E are the energies of tunneled electrons

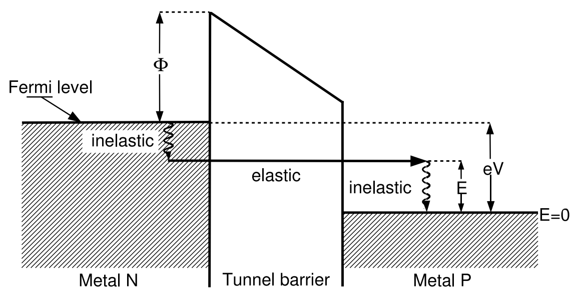

Fig. 3. The energy scheme of normal-metal – insulator – normal-metal TJ at external bias voltage V. Elastic electron tunneling process at the energy E, marked by an arrow, is followed by inelastic processes accompanied by emission of quasiparticles with total energy E and $eV - E$ in metal P and metal N, respectively. The maximum energy of generated quasiparticles is $\hbar\omega_0 = eV$.

counted from the Fermi level of metal P. As can be deduced from Figure 3, the process of elastic electron tunneling at energy E is accompanied by inelastic electron process(es). These occur as the electron, after passing the barrier, appears in an excited state with excess energy E above the Fermi level of metal P. It therefore has to be brought into the equilibrium state via inelastic collisions generating non-equilibrium quasiparticles, e.g. phonons, with total energy E. Analogously, as the empty state (non-equilibrium hole) has been created below the Fermi energy in the metal N it has to be filled by the electron from higher energy level. This is accompanied by inelastic electron process(es) in metal N with total energy $(eV - E)$. Thus, based in equation (1) and considering that the energy of all inelastic electron processes in electrodes will be as result converted to heat, one can write

$$P_{pos} = \int_{-\infty}^{+\infty} En(E)dE \tag{2}$$

$$P_{neg} = \int_{-\infty}^{+\infty} (eV - E)n(E)dE, \tag{3}$$

so that, for the power dissipated in the whole TJ we have

$$P_{pos} + P_{neg} = VI. \tag{4}$$

As can be seen from Figure 2 and its inset, experimental data are in excellent quantitative correspondence with equation (4) yielding the proof that the CT experiment measures the energy dissipated in TJ electrodes in absolute units. The profile of P-V curves from Figure 2. can be explained using equations (2, 3) as follows. At sufficiently low temperatures ($k_BT \ll eV$) for E out of the range $\langle 0, eV\rangle$ $n(E)$ can be regarded as zero, while in the range $\langle 0, eV\rangle$ it can be approximately expressed by $n(E) = \rho_N(E - eV)\tau(E, eV, z, \phi)\rho_P(E)$ (ρ_P and ρ_N are the densities of states of the positive and negative electrode, respectively, ϕ is the output work of electrodes, τ is the transmission barrier probability). In the low-voltage limit ($|eV| \ll \phi$), assuming normal-metal electrodes, $n(E)$ can be considered as constant in the range $\langle 0, eV\rangle$ yielding $P_{pos} = P_{neg} = VI/2$. At higher bias voltage the exponential behavior of $n(E)$ has to be taken into account, as the energy dependence of $n(E)$ in equations (2, 3) will be preferably governed by $\tau(E, eV, z, \phi)$ which is an exponential function of E. As V increases, the states with energy slightly lower than $E = eV$ give ever more important contribution to the tunnel current. For P_{pos} the contribution from these states is enhanced due to the multiplication of $n(E)$ by E, while for P_{neg} the situation is just the opposite, due to the term $(eV - E)$. This explains the steeper growth of P_{pos} in comparison to P_{neg} with increasing V, and the tendency of P_{neg} to saturate at highest V, where the dominating contribution to the tunnel current originates just from the states from the region close below $E = eV$.

The qualitative difference between P-V and P-I characteristics explains the fact that while a change of tunnel current at constant bias voltage changes the mutual distance between TJ electrodes and therefore governs properties of the tunnel barrier characterized by τ, the change of bias voltage, in addition, influences the energy scheme of TJ (and defines the energy window for inelastic electron processes).

As follows from the discussion above, the unique property of CT studies is that in contrast to I-V characteristic studies, where the tunnel current is measured as a common property of both TJ electrodes, studies of P-V characteristics yield information about energy processes for each of the TJ electrodes separately. Due to this, the CT experiment is sensitive to all kinds of physical processes causing an energy transfer from/between TJ electrodes, such as light emission caused by tunnel currents [14] or selective extraction of "hot" electrons from normal-metal to superconductors [7]. However, if there is no transfer energy from the tunnel structure, the coupling between I-V and P-V dependencies via equations (1–3) implies the equivalence of both approaches. Therefore we claim that *careful* studies of P-V characteristics can be used for derivation of energy-spectroscopic information analogously to "classical" tunneling spectroscopy based on measurements of dI/dV or d^2I/dV^2 [8]. [We note that at sufficiently low temperatures ($k_BT \ll eV$) the equivalence $dI/dV = (1/V)dP_{pos}/dV$ takes place, as we showed elsewhere [15].]

The results presented clearly indicate a different nature of charge injection in PCs and TJs. While in ballistic PCs, the energy gained due to the acceleration in the field region of the contact is expected to dissipate only in the electrode into which the electron is injected, in TJs heat generation in *both* electrodes takes place. For an illustration, following the energy diagram in Figure 3, the charge injection in PCs can be imagined as "resonant tunneling" directly from the Fermi level of metal N into the state with energy eV above the Fermi level of metal P so that the whole energy eV is dissipated in the metal P only. If we characterize the asymmetry of heat production by $A = (P_{pos} - P_{neg})/(P_{pos} + P_{neg})$ then for the ballistic PC limit $A_{PC} = 1$, while for the case of the normal-metal – vacuum – normal-metal TJ in the low-voltage limit $A_{TJ} = 0$. For the contacts with the "ballistic PC–TJ crossover behavior" (e.g. like the crossover from metallic to TJ behavior for normal-metal – superconducting microconstriction contacts introduced by Blonder, Tinkham and Klapwijk [16]) the value of A should lie between A_{PC} and A_{TJ}. This offers an experimental possibility for the determination of mutual contribution of *tunneled* and *ballistically injected* carriers in "crossover" contacts. Of course, such type of experiments require a non-vacuum tunnel barrier and a consequent consideration of the thermal coupling between the tip and the sample.

From the point of view of possible applications of the CT experiment (namely in the field of design and development of electronic devices) we would like to emphasize that a TJ represents a source of thermal gradients, especially at high bias voltage and "non-constant" behavior of $n(E)$. In addition, non-equilibrium charge carrier distribution due to tunneling and consequent generation of quasiparticles (e.g. phonons), which can be effectively studied by the CT experiment, play a fundamental role in many applications, especially at very low temperatures and in a small electronic devices where thermalization of the electron system presents a fundamental problem [17]. For instance, electrical conduction in impurity bands can occur via phonon-assisted tunneling between localized states, and therefore generated non-equilibrium phonons can be a reason for an additional channel of electrical conduction. Analogously, the spin relaxation rate due to phonon scattering, which plays an important role in spintronic applications [18], can be influenced by non-equilibrium phonons generated due to the tunneling/injection of spin polarized charge carriers. Thus the heat generation/sinking accompanying the charge tunneling is a rather complex task which can not be sufficiently described only by the total dissipated power and corresponding overheating and/or generation of thermal gradients, as generated quasiparticles, whose energy window is defined by applied bias voltage, can drive physical processes in each of the TJ electrodes. We believe that the CT experiment represents a powerful tool suitable for studies of energy processes accompanying a charge injection, that according to its nature and its capability to be extended to a scanning probe microscopy technique will provide answers to many open questions important for applications in electronics (including nanoelectronics).

In summary, the principle of novel calorimetric tunneling experiment has been proposed. Using this experiment we have measured the heat generated in normal-metal – vacuum – normal-metal TJ. Results of the measurements show that the mutual redistribution of the power dissipated in TJ electrodes is symmetrical in the low voltage limit, however, it shows a marked asymmetry at higher bias voltage. Moreover, they indicate that the generated heat represents the energy of non-equilibrium quasiparticles coming from inelastic electron processes which accompany the process of electron tunneling. According to its nature the CT experiment is sensitive to all kinds of physical processes causing an energy transfer from/between TJ electrodes and it can be extended to spatially resolved techniques by means of scanning probe microscopy. It can also be applied for energy spectroscopy of electrically conductive solids, studies of thermoelectric effects in tunnel structures, as well as in development of TJ containing devices whose functionality is influenced by temperature gradients or by generation of non-equilibrium charge carriers and/or non-equilibrium quasiparticles, such as electronic microrefrigerators based on a NIS (or similar) tunnel structures or Coulomb blockade thermometers.

We thank Prof. M. Meißner for the opportunity to perform our first experimental attempts to measure heat generation in VBTJs in the Heat Capacity Laboratory at Hahn-Meitner-Institut Berlin and for his useful discussions and recommendations on the calorimeter design. This work was supported by Centre of Low Temperature Physics operated as the Centre of Excellence of the Slovak Academy of Sciences under Contract No. I/2/2003 and by the VEGA grant No. 2/4050/04.

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