

Filming the thermal death of electrons in matter

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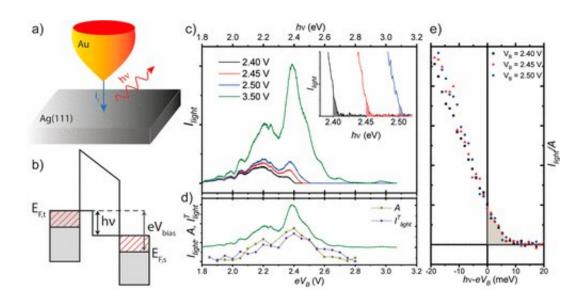


Figure 1. (a) Schematic representation of the experiment: a tunnel current flows from a Au STM tip to a Ag(111) surface exciting plasmons, the radiative decay of which leads to photon emission. (b) Level diagram showing that the width of the energy window of possible initial and final states of an inelastic tunnel process exciting a plasmon of energy hv is $eV_B - hv$; i.e., for low photon energies, more inelastic transitions contribute to the emission. For photon energies higher than the bias voltage, inelastic processes linking occupied states in the tip and empty states in the sample become impossible. (c) Tunnel electroluminescence spectra recorded at 4.9 K with a bias voltage of 3.5 V, where all the relevant plasmonic cavity modes can be accessed by inelastic processes, and at lower voltages (2.4–2.5 V), demonstrating the suppression of intensity at photon energies larger that the applied bias. Inset: Zoom into the emission edge. The overbias emission tail is shadowed. (d) Comparison between the voltage dependence of the overbias amplitude (A, i.e., the light intensity at the cutoff) and total integrated emission (I_{light}^{T} , i.e., integrated light intensity at



energies larger than the cutoff) with the fully developed spectra at 3.5 V. (e) Normalization of the emission edge spectra at different voltages by their respective amplitudes, A, makes the spectra voltage independent. Credit: DOI: 10.1021/acs.nanolett.1c00951

It is well known that an electric current increases the temperature of the material through which it is conducted due to the so-called Joule effect. This effect, which is used daily in domestic and industrial heaters, hair dryers, thermal fuses, etc., occurs because the new electrons injected into the material cannot go to the lower energy states because those are already occupied by the electrons of the material and therefore they must start their journey with relatively high energies. These electrons are called hot carriers. However, as they move through the material, hot carriers lose energy through collisions with other electrons and atoms in the solid. The process by which this lost energy is translated into thermal energy and, therefore, into an increase in temperature, is known as thermalisation of hot carriers.

It should be noted however that this well-known effect takes place for very high electron fluxes, which can reach billions of electrons per second in electronic conventional devices. Therefore, it reveals information about the collective behavior of electrons, but how long it takes each of them to lose their <u>energy</u> is a generally difficult question to answer experimentally.

In an article published in *Nano Letters*, a group of Spanish researchers has proposed a new method to explore the thermalisation of hot carriers with temporary resolution of billionths of a second. The work, which results from a collaboration between the Autonomous University of Madrid, IFIMAC, the Madrid Institute for Advanced Studies in Nanoscience (IMDEA Nanociencia), the Donostia International Physics



Center (DIPC) and the University of the Basque Country (EHU), used a scanning tunnel microscope to inject electrons into a silver surface at a rate thousand times lower than that corresponding to operating currents in standard devices. The researchers examined the energy distribution of the emitted light at the junction in response to electron injection.

A naïve view of the law of conservation of energy would imply that photons should not be emitted with energies greater than the voltage applied to the junction: The experiment, on the contrary, shows that although the number of photons with energies greater than the applied voltage is very small, it is not completely zero. In its work, the consortium, led by Prof. Roberto Otero, explains this phenomenon as the result of taking into account the <u>temperature</u> of the electron cloud of the solid, and allowed the researchers to extract this temperature from the energy distribution of the photons with energies above the voltage.

This analysis shows that the temperature of the electron cloud and that of the material itself do coincide for high temperatures and low currents. However, as the current is increased, the estimated electronic temperature increases above the sample temperature. The authors rationalize this behavior taking into account that, by increasing the current, the average time between the injection of consecutive electrons decreases. When this time is less than the time corresponding to the thermalisation of hot carriers, the second electron injected notices the electron cloud temperature is higher than the one of the sample, because the energy of the first electron has not yet been completely dissipated. If the injection of the second electron results in the emission of light, the energy distribution of the light with energies above the voltage will reflect the temperature of the electron cloud at the time of injection. In this way, by measuring the emission of light with energies above the voltage at different currents it is possible to follow the speed with which the thermalisation process takes place.



The study clarifies the nature of photon emission above the applied voltage and shows how this fact is perfectly consistent with current scientific knowledge. Additionally, it offers a new way of measuring the electronic temperature of solids via scanning tunnel microscope with atomic spatial resolution. And it offers a new tool to study the thermalisation processes of hot carriers one at a time. For all these reasons, the authors are confident that this work is essential for the design and characterization of nanoscale thermal and luminescent devices, and could have important implications for the design of nanometer catalysts for different chemical reactions, or the manufacture of nanometer lasers that could work with extraordinary low pump powers.

More information: Alberto Martín-Jiménez et al, Electronic Temperature and Two-Electron Processes in Overbias Plasmonic Emission from Tunnel Junctions, *Nano Letters* (2021). DOI: 10.1021/acs.nanolett.1c00951

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