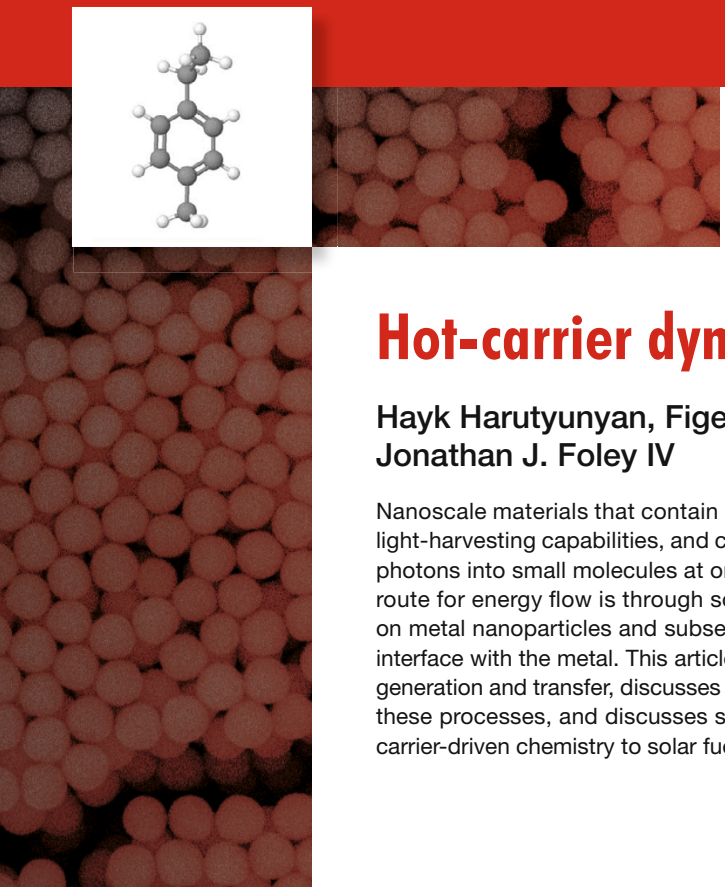

# Hot-carrier dynamics in catalysis

Hayk Harutyunyan, Figen Suchanek, Robert Lemasters, and Jonathan J. Foley IV

Nanoscale materials that contain metallic components can be designed to have excellent light-harvesting capabilities, and can also be used to direct the flow of energy from incident photons into small molecules at or near the surface of metal nanoparticles. One promising route for energy flow is through so-called hot charge carriers, which are optically excited on metal nanoparticles and subsequently transferred to molecules/materials that share an interface with the metal. This article provides an overview of the fundamentals of hot-carrier generation and transfer, discusses both theoretical and experimental means for interrogating these processes, and discusses several potential societally important applications of hot-carrier-driven chemistry to solar fuels and sustainable chemistry.

## Introduction

The understanding of the dual particle-wave nature of light and matter was a watershed moment in our basic understanding of how energy flows in the universe; this has been leveraged extensively in advancing experimental and theoretical tools to elucidate, and even control, the flow of energy in chemical and materials systems where light and matter converge.[1] Hot-carrier-driven catalysis represents a curious trinity of energy flow—incoming light imparts energy into electronic degrees of freedom, which in turn, impart energy into nuclear degrees of freedom. Not only is this interplay between photonic, electronic, and phononic/nuclear energy fundamentally interesting, it shows potential to enable technologies that are capable of efficiently harvesting energy from solar photons or using low-intensity light sources to drive photochemical transformations under mild conditions, which are both important from the point of view of sustainability.[2–4]

Here, we focus on metal nanoparticles as sources of hot carriers. Such nanoparticle systems may derive their efficient hot-carrier generation properties from intrinsic optical resonances (such as localized surface plasmon resonance, LSPR)[2–4] or emergent optical resonances (such as scattering mediated absorption, SMA).[5,6]

## Hot-carrier generation and transfer

Hot-carrier transfer from metal nanoparticles to molecules to initiate photocatalytic reactions is an intrinsically multiscale problem—the constituents span multiple length scales and the energy-transfer events span multiple time scales. On the shortest time scale (~10 fs), electromagnetic energy flows into the electronic degrees of freedom of the nanoparticle(s) and molecule(s) subject to their mutual physio–chemical interactions; these interactions may have a small or large impact on the electronic degrees of freedom depending on the identity of the metal and the molecules.[4] Plasmonic excitation and decay also occur on this time scale.[2–4]

A slightly longer time scale (tens to hundreds of fs) sees an evolution of the hot-carrier distributions, including thermalization through electron–electron scattering, decay through electron–phonon scattering, and transfer to adsorbates.[4,7–9] The transfer of charge from the metal to the molecule deposits energy into the degrees of freedom of the molecule, but also fundamentally changes the potential energy surface that governs the nuclear dynamics of the molecule. Nuclear motion of the adsorbate molecules evolves on the potential energy surface(s), corresponding to excited or ionized states of the adsorbate molecule; often the interatomic forces that govern the molecular motion upon excitation and ionization are considerably changed

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and may include different or more facile pathways. Nuclear motion typically occurs on slower time scales (~hundreds to thousands of fs), though theoretical studies have suggested ultrafast (~30 fs) nuclear dynamics following plasmon-induced electron transfer may occur in water-splitting reactions.[8] The time scales involved are illustrated schematically in **Figure 1**.

## Theory and simulation of hot-carrier-driven catalysis

It is interesting to imagine what a comprehensive predictive computational model of hot-carrier-driven catalysis would look like. In our view, upon specification of the materials, the geometry of the nanostructure and adsorbed molecule(s), and details of the illumination source, such a tool would predict the flow of electromagnetic energy into the electronic degrees of freedom of the nanoparticle and molecule subject to their mutual physio–chemical interactions. It would also subsequently predict the electronic motion of the excited system, and the nuclear motion to allow inference of the potential reactive probabilities and pathways. At this point, there exist no such routine comprehensive theoretical/computational approaches; however, one can simulate each subprocess on its own temporal time scale and gain some insights into the integrated processes. This suggests that continued progress, particularly along the direction of multiscale theory and simulation, will yield theoretical/computational tools that can provide insights and aid in the design of systems for performing desired hot-carrier-induced catalysis.

The photonic problem (shortest time scale), which is the interaction between light and the nanoscale components (i.e., solving for the electric field in the vicinity of the nanoparticle, $\vec{\mathbf{E}}_{NP}$ in Figure 1), can be treated within classical electrodynamics. A wide variety of analytical and numerical techniques can be deployed to accurately simulate the flow of electromagnetic fields in the vicinity of nanomaterials. Analytical methods such as Mie theory[10] and the T-Matrix scattering method[11] may be deployed if the nanoscale constituents can be modeled as lone spheres or as arrangements of spheres with non-overlapping boundaries. For arbitrary geometries, the discrete dipole approximation[12] and finite element method[13] may be used to solve the behavior of the electromagnetic fields in the frequency domain, and the finite-difference time-domain method can be used to solve the flow of the electromagnetic fields in the time domain.[14] These techniques are critical for providing insights into how the nanoscale structure harvests and directs the flow of optical energy, which will influence the generation of hot carriers.

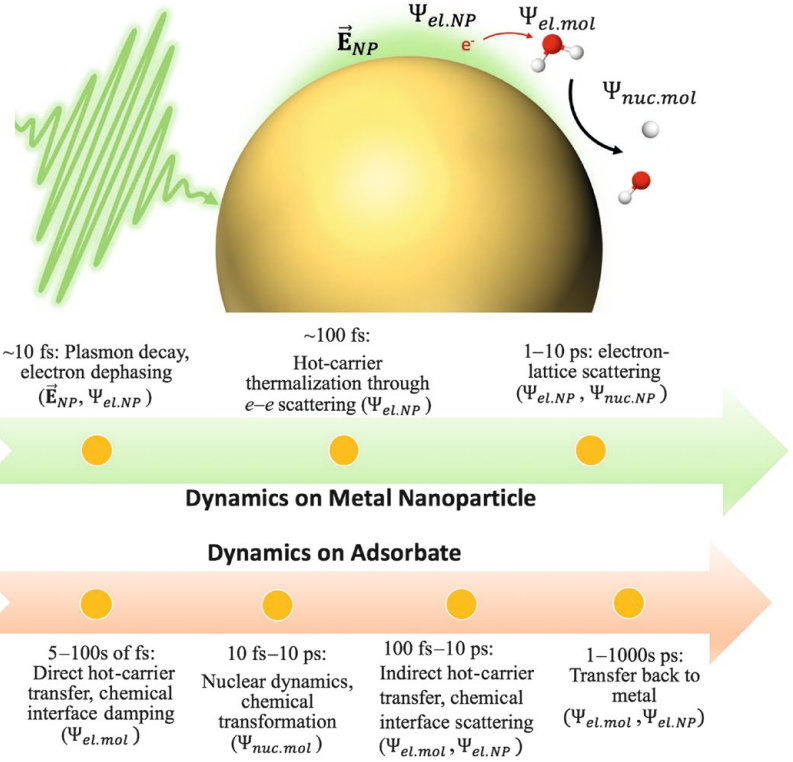


**Figure 1.** Schematic of hot-carrier driven catalysis with metal nanoparticles, with time scales and key quantities involved in each subprocess. $\vec{\mathbf{E}}_{NP}$ represents the electric field (i.e., photonic degrees of freedom) in the vicinity of the nanoparticle, $\Psi_{el.NP}$ ($\Psi_{el.mol}$) represent the electronic wavefunctions, electronic degrees of freedom, of the nanoparticle (molecule), $\Psi_{nuc.NP}$ ($\Psi_{nuc.mol}$), represent the nuclear wavefunctions, nuclear degrees of freedom, of the nanoparticle (molecule), and $e^-$ represents a hot electron in transit from the nanoparticle to the molecule.

All things being equal, nanoparticles that can more efficiently harvest and concentrate incident optical energy will see higher rates of hot-carrier generation,[15] and nanoparticles with longer-lived optical resonances will see more persistent hot-carrier dynamics.[6] Modeling hot-electron dynamics (intermediate time scale) and transfer requires a quantum mechanical treatment of the electronic degrees of freedom of the metal nanoparticle (NP) and adsorbate molecule ($\Psi_{el,NP}$, $\Psi_{el,mol}$), respectively. A surge in computational advances have increased the efficiency of electronic structure methodologies, density functional theory (DFT) in particular, and have enabled detailed investigations of the electronic distributions and their dynamics (through the real-time implementation of time-dependent DFT) in small (~1 nm) metal nanoclusters,[8] though application of *ab initio* methods to larger nanoparticles remains intractable. A variety of semiempirical approaches, including the so-called Jellium model[6,15,16] and DFT tight binding[17] have been employed to study larger nanoparticle systems, since they effectively coarse-grain over core electronic degrees of freedom. These methods are still limited by relatively steep (~quartic) scaling with the system size.

Finally, progress has been made toward the treatment of nuclear dynamics on (perhaps multiple) excited-state or ionized potential energy surfaces, including the use of Ehrenfest dynamics[8] and fewest-switches surface hopping[9] in conjunction with time-dependent DFT. Also, impressive

advances in wavefunction embedding theories have permitted the accurate mapping of relevant molecular potential energy surfaces with inclusion of physio–chemical interactions with metal nanoparticle surfaces.[18] Wave-function embedding approaches, more than DFT or semiempirical approaches, offer a more systematic route to including high-level electronic correlation effects, which may be significant for accurately capturing the shape of excited-state potential energy surfaces that govern the reactivity in these processes.

## Experimental characterization of hot-carrier-driven catalysis

Experimental realization and characterization of efficient hot-carrier platforms (i.e., plasmonic and other optical nanostructures) encompass a wide variety of sample fabrication approaches and experimental techniques. Typically, the samples used for characterization of hot-carrier generation can be categorized into two groups—colloidal and nanofabricated. The colloidal approach has the advantage of using wet chemistry methods for tailoring material composition and structure of the nanoparticles. Using this approach, high-quality nanoparticles with good crystalline structure can be obtained.[19] Using more sophisticated techniques, more complex hybrid nanoparticles with desired electronic and optical properties can be synthesized.[20] On the other hand, nanofabrication approaches offer the ability to control the size and shape, thereby imparting exquisite control of optical energy flow and confinement, owing to the flexibility of the lithographic and material deposition techniques.[21] Using this approach, arbitrary shapes of plasmonic platforms can be fabricated, which potentially allows access to new hot-carrier generation mechanisms. For example, the ability to fabricate tapered nanostructures has allowed the study of hot-carrier generation in adiabatically focused plasmonic fields and the shaping of the momentum distribution of the hot carriers.[22,23]

Various optical experimental approaches have been used to study the processes of photo-induced hot-electron generation and injection in the temporal, spatial, and frequency domains. Because of the fleeting nature of hot carriers, it is usually necessary to employ time-resolved experimental techniques in order to observe associated short-lived phenomena. Pump-probe transient spectroscopy offers ultrafast temporal resolution only limited by the pulse duration of the excitation source (i.e., from 10 fs to 100 fs).[24] This allows access to the fast dynamics of electron excitation and relaxation in nanostructures.[25,26] In a typical experiment, the pump pulse excites the LSPR, which leads to the production of energetic carriers in the system.[21,27] The broadband probe pulse can subsequently probe the reflectivity of the system and changes in the dielectric constant of the material caused by the excited hot carriers in the system.[28]

Using these techniques, effects such as field localization and enhanced generation of hot electrons in nanometric gaps have been demonstrated.[21] Resonant hot-carrier generation can be understood in terms of Landau damping, where the localization on the characteristic length scale in real space creates large field gradients leading to resonant dephasing of the plasmonic and free carrier intraband absorption in the metal.[29] The length scale in this process is given by the band curvature of the conduction band (i.e., the Fermi velocity) and can be compared to the Kreibig term of the plasmonic damping of small nanoparticles.[30] Furthermore, ultrafast transient absorption studies can uncover the mechanisms of hot-carrier generation and injection in hybrid metal–semiconductor nanocomposite particles.[31] In such studies, typically, the probe pulse monitors the time-resolved absorption in the semiconductor at the corresponding bandgap energies and thus any transient changes in absorption can be indicative of hot-carrier injection across the metal interface. Lian et al. used a similar approach and reported direct decay of plasmons in colloidal metal nanoparticles into an excited carrier in an adjacent semiconductor, beating the conventional Fowler limit for a two-step generation and charge-transfer mechanism in a typical Schottky-injection scheme.[20] Pump-probe spectroscopy also enables the study of the spatial distribution of excited hot carriers. Using this approach, the diffusion of the generated carriers can be traced in the samples and information about the spatiotemporal dynamics at different scales can be extracted.[23,32,33]

In the frequency domain, hot-carrier generation and transfer can also be studied using emission spectroscopy techniques. The emission spectrum can be used to gather information about the hot-electron energy distribution and dynamics. Various physical processes have been used to describe the hot-carrier distribution in nanoscale systems. Most commonly, photoluminescence (PL) spectroscopy is used to characterize hot-electron generation in plasmonic structures.[34] In such studies, pulsed excitation laser sources are typically used to access the multiphoton regimes of the PL emission. However, it has recently been demonstrated that emission excited by continuous wave excitation can also produce invaluable information about the hot-carrier distribution in the nanosystems.[35] Early studies on gold nanostructures attributed the emission at different spectral ranges to distinct physical processes.[36] Whereas the two-photon absorption-induced luminescence at visible frequencies was thought to arise from intraband transitions—the near-infrared emission was assigned to intraband absorption (i.e., hot-electron generation). The intraband transition process, which scales linearly with excitation power, is facilitated by plasmonic confinement, making the resonant free carrier absorption possible in noble metals. This was further developed in later studies where a continuous variation of the emission power exponent was observed across the PL spectrum.[34,37] Such noninteger values of the PL power exponent were assigned such that the emission throughout the whole spectrum has the same origin (i.e., the radiative recombination of thermally equilibrated hot-electron gas excited via intraband absorption). Later studies added novel dimensions to our understanding of hot-electron-mediated emission by invoking other mechanisms for the observation of the broadband signals. Raman scattering has been used to explain some of the spectral features of plasmon-enhanced emission.[35,38] In such a process, inelastic scattering occurs by exciting a conduction-band electron and a lattice phonon. Thus, the hot-electron

energy distribution above the Fermi energy is imparted onto the Raman scattering spectrum, similar to PL emission.

## Hot-carrier-driven photocatalysis

The strategy for hot-carrier-driven photocatalysis fits broadly within the push for advancing sustainable paradigms in chemistry.[2–4,39] Most directly, a tremendous amount of progress has been made toward developing photocatalytic and photoelectrochemical systems that leverage hot-carrier transfer to facilitate energy-intensive chemical reactions that produce chemical fuels (e.g., water splitting to $H_2$ and $O_2$[4,8,40–42] or $CO_2$ reduction to $CH_4$[43]) or important feedstocks for fuels, including $CO_2$ reduction to CO (**Figure 2**).[4,39] Because plasmonic and other resonant metal nanoparticles are extremely efficient light harvesters, these reactions can be initiated with relatively low illumination conditions (~100 mW/cm$^2$, similar to solar illumination), which suggests the possibility of utilizing such paradigms for solar fuels.

As one concrete example, Moscovits and co-workers reported plasmonic gold nanorod-based solar water-splitting devices capable of yielding $5{\cdot}10^{13}$ $H_2$ molecules/s/cm$^2$ device coverage under solar illumination (around $1{\cdot}10^{18}$ visible photons/s/cm$^2$ ).[39] Under 3-sun illumination, the same system yielded about 2.8 mmol $H_2$/h/g of catalyst.[39] A similar yield (2 mmol of $CH_4$/h/g of catalyst) was reported by Garcia and co-workers[43] using heterostructures involving plasmonic gold, copper, and titanium oxide whose selectivity for $CO_2$ over $CH_4$ was enhanced by hot-carrier injection from gold to copper. While these and other studies are exciting, the area of hot-carrier driven solar fuel generation remains relatively immature, and it is difficult to project cost competitiveness from laboratory-scale studies to date.[39] Important cost benchmarks include the price to generate these products using fossil fuels; for example, fossil-fuel-driven production of $H_2$ via steam methane reforming is estimated to cost around USD \$1.40/kg $H_2$ and production of CO from partial oxidation of fossil fuels is estimated to cost around USD \$0.19/kg CO.[39]

It is our view that continued research efforts, both in fundamental and applied domains, should keep in mind several important goals, including (1) increasing efficiencies of hot-carrier driven reactions (in terms of photon-to-chemical yields or chemical-yields to catalyst-mass, or both); (2) realizing such reactions using little or no precious metals; and (3) increasing the durability and lifetime of such catalytic systems, which can all positively impact cost competitiveness over the lifecycle of these systems. A critical economic analysis accompanying these efforts could be illuminating for the overall prospect of utilizing hot-carrier driven photochemistry on an industrial scale.

Besides solar fuels, hot-carrier-driven catalysis can also be used to increase the sustainability of chemical reactions more broadly, for example, by lowering the energy requirements of chemical reactions that are traditionally run at high temperatures, or by improving the selectivity of reactions that may undergo side reactions or degradation under high-temperature conditions. For example, plasmon-induced hot-electron injection from gold/palladium heterostructures has been suggested to efficiently activate the C–Br bond to facilitate a number of Suzuki coupling reactions, which are widely used in pharmaceutical and industrial chemistry, and has resulted in twofold improvements in yields when compared to their thermally activated counterparts.[44] Recently, hydrogenation of alkynes to alkenes and alkanes was demonstrated using hot electrons from periodic gold/platinum heterosurfaces, where hot-electron generation results from surface plasmon polariton excitation on gold, and subsequent hot-carrier transfer and activation of alkyne triple bonds is facilitated by the platinum.[45] Such studies highlight the vast potential in functionality that can be realized by creating heterostructures with one or more components that serve as hot-carrier sources.

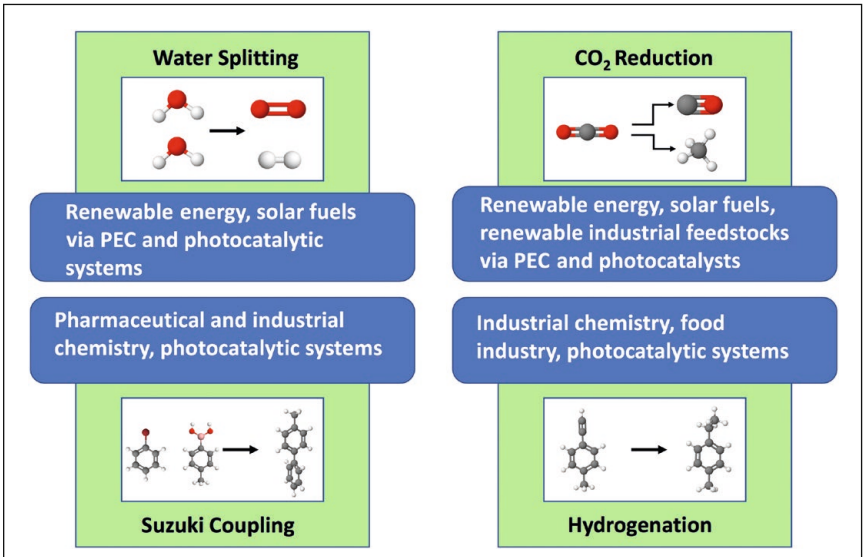


**Figure 2.** Illustrations of hot-carrier-mediated chemistry for solar fuels (water splitting and $CO_2$ reduction) and for ubiquitous synthetic steps utilized across many industries (Suzuki coupling and hydrogenation reactions). Note: PEC, photoelectrocatalysis.

## Summary

Hot-carrier dynamics in catalysis encompasses photophysical and photochemical processes across a diverse set of length- and time scales, making this topic extremely rich from a phenomenological point of view. Theorists and experimentalists have been drawn to study the rich behavior of hot-carrier dynamics both in pursuit of deeper fundamental understanding, and for the potential of controlling and optimizing hot-carrier dynamics to facilitate new and sustainable strategies for chemical transformations. The progress made in fundamental understanding and in realization of useful chemistry involving hot-carrier dynamics is promising and should be continued. It should also be accompanied by substantial efforts to critically evaluate and advance their potential efficiency, cost-competitiveness, and scalability.

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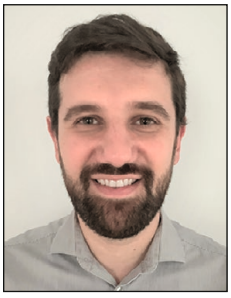

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