Infrared Photoemission from a Pure Gold Surface: Validation of the Lucky-Electron Model

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We have observed a strong dependence of the photoelectron current with the sample temperature in a midinfrared laser-surface interaction experiment. We show that classical mechanics is able to interpret this unexpected phenomenon. This is demonstrated with a "lucky-electron" simulation based on a phase-matching condition between the temperature dependent mean electronic collision time and the laser electric field. This opens ways for measurements of temperature dependent effective scattering rate in the midinfrared.

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For this visible range of the electromagnetic spectrum, great progress has been made in recent years in the understanding of the interaction of a laser field with pure metallic surfaces. This is mainly due to femtosecond laser studies of hot electron dynamics [1,2] and to the development of theoretical quantum models based on the resolution of the time-dependent Schrödinger equation [3].

If everything is far from being perfectly understood, the situation is much more embryonic when the laser wavelength is in the midinfrared where, surprisingly, only a very small number of photoemission experiments has been performed. In particular, it has recently been observed that for very moderate intensities ($\sim 10^6$ W/cm^2), the interaction of midinfrared (~ 10 μm) short laser pulse (~ 2 ps) with a gold surface was causing an unexpected intense electron emission and a corresponding "hot" electron spectrum (up to a few eV's) in conditions free of lattice heating by excited electrons [4]. Among the possible descriptions of this phenomenon, a simple model, first introduced by Seitz [5], based on classical dynamics approach and considering the conduction electrons as a free electron gas in the presence of random collisions, has been shown to be suitable to reproduce the experimental data. Its main assumption is that, as a result of the collision, an electron suddenly changing the direction of its momentum can be accelerated (or decelerated) depending on the phase of the laser field at that moment. As in the Drude model, it is not necessary to have a precise understanding of the detailed mechanism of the collisions.

The purpose of the present work is to test this model which predicts that the amount of photocurrent should depend on the mean collision time (MCT) τ via the quantity $\omega\tau$, where ω is the laser frequency. The condition $\omega\tau = \pi$ is the phase-matching condition which is of importance in the optimization of the energy absorption process. Because, in gold at room temperature, the MCT is of the order of 10 fs [6], this condition is fulfilled for laser wavelength of the order of 7 μm. We have then performed a photoemission experiment at 7.6 μm, varying the MCT. This can be achieved by changing the sample temperature [7]. Our experimental results will be compared directly with the simulations.

Within this framework, let us consider a free electron driven by the laser ac electric field $E_0 \sin(\omega t)$ but undergoing oscillations with a mean time τ. If $\omega\tau \gg 1$, the electron oscillates nearly freely in the field and the energy change in a half cycle is compensated during the next one. This condition is achieved in the visible or in the near infrared range ($T_{\text{opt}} = \frac{2\pi}{\omega} \sim 2.5$ fs) of the spectrum and this well known regime is the so-called "multiphotonic regime" [8]. On the contrary, when $\omega\tau \ll 1$ (low frequency limit), the electrons undergo many collisions during an optical cycle and the energy gain is still very small because the acceleration is inhibited.

When $\omega\tau = \pi$, the situation is very specific because an electron has the possibility to perform on average 2 collisions per optical cycle. It has then the possibility to be accelerated twice and, by changing its momentum right in the middle of the optical cycle, it will be able to gain a velocity $\alpha = \frac{2eE_0}{m\omega}$ as it is explained explicitly below. Thus, if we define N as the maximum number of collisions during a given pulse duration, the velocity gain will be $N\alpha$. In this case, an electron could gain an amount of energy larger than the work function ($W_s \sim 5.5$ eV) of the metal. This situation, is, of course, ideal because it describes the story of an electron which has been continuously accelerated all along the laser pulse. It turns out the probability of such a very efficient sequence of alternating velocity directions is extremely low. It can be estimated in this simple one-dimensional model as $(\frac{1}{2})^N$. For a 2 ps pulse duration and 10 fs as MCT, $N = 200$ and this probability is of the order 10^{-60}. It is more realistic to simulate individual electron trajectories by including a random statistics on the time of the collisions vs the phase of the laser field, as they can take place whenever in the cycle. An electron which gains a kinetic energy larger than the work function and then has the possibility to escape into the vacuum, is called a "lucky electron."

To present the model more explicitly, let us consider the one-dimensional case for the sake of simplicity. The time of the nth collision is $t_n = t_1 + (n-1)\tau$ where t_1 corresponds to the first collision. If we suppose that collisions are isotropic, following a trajectory is equivalent to drawing a random sequence of numbers (ϵ_n) with two possible values: $+1$ if the collision is forward and -1 if the collision is backward. In the presence of the laser field, the electron velocity reads between two collisions: $v_{n+1} = v_n + \frac{\alpha}{2}\epsilon_n[\cos(\omega t_{n+1}) - \cos(\omega t_n)]$. If we consider an electron starting at $t = 0$ with v_F as the Fermi velocity, its velocity will be after N collisions: $v_N = v_F - \alpha\epsilon_0 \sin^2(\omega t_1) - \alpha \sin(\omega\tau/2)[\sum_{n=1}^{N-1} \epsilon_n \times \sin(\frac{2n-1}{2}\omega\tau + \omega t_1)]$. Note that if $\omega\tau = \pi$ (perfect phase matching), if the sequence is alternatively ± 1, and if $t_1 = 0$ (the electron starts when the electronic field is zero), the maximum velocity gain $(N\alpha)$ is, as already mentioned, reached. In a three-dimensional model, the expression is very similar except that ϵ_n is replaced by $\cos(\theta_n)$, the angle between the velocity and the laser electric field.

Considering only electrons at the surface of the Fermi sea, the kinetic energy distribution (energies $> W_s$) at the end of a 7.6 μm, 2.5 ps, 5 MW/cm^2 laser pulse has been simulated. The result is shown in Fig. 1 (inset). This spectrum has been obtained by Monte Carlo simulation using 10^8 particles. The MCT has been set to 10 fs (room temperature) and was not allowed to vary. Different scattering time distributions (square or Gaussian) with various widths have been tested without significant qualitative differences: only the absolute electron number is changed. Because 7.6 μm corresponds to $T_{\text{opt}} = 24$ fs, there is on average 2.4 collisions per cycle not far from the optimum value of 2 collisions per cycle. We also present in Fig. 1 the calculated photocurrent as a function of the laser intensity ($I \sim E_0^2$). It increases with the intensity because the energetic "jumps" increase with I which entails a decrease of the required number of events leading to ionization.

We now turn to the discussion of the actual realization of such a scenario in an experiment. The IR-FEL at the CLIO facility at LURE (Orsay, France) delivers series of laser pulses of duration about 2 ps. In our experiment, the wavelength has been set to 7.6 μm and the intensity was in the MW/cm^2 range. The ultrahigh-vacuum chamber ($P < 10^{-9}$ mbar) contains a joule heating filament and a cooling nitrogen circulation located behind the sample. The temperature can be varied between -160 °C and 450 °C. The electron signal is detected with a two-microchannel plate system coupled to a fast oscilloscope working in counting mode. We record simultaneously the photoelectric current and the pulse energy for each laser shot. The incidence angle is 70° and the laser is p polarized on the sample through a ZnSe window. The gold (100) monocrystal sample has been very carefully prepared. It has been mechanically polished reaching a flatness of $\sim\lambda/20$ before an electrochemical polishing. Under UHV conditions, up to 10 *in situ* Ar^+ ion bombardment cycles separated by heating cycles (450 °C) were performed before each experiment. We have verified that in our experimental conditions the electronic signal was not correlated with any sign of desorption process for the whole range of temperatures by a careful verification of the pressure in the chamber.

The experimental results (same conditions as in Fig. 1) are shown in Fig. 2. The photoelectron signal is observed as a function of the sample temperature which was continuously increased (or decreased). A bell-shaped curve is observed. We have verified that the curve was flat at temperature above 120 °C and that the average laser power was constant over the whole acquisition duration.

In order to check our experimental setup we have also performed the experiment in the same conditions, by

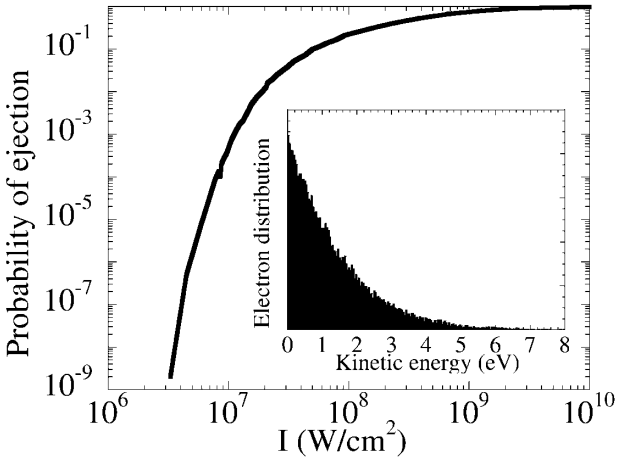

FIG. 1. Simulation of the probability of ejection of an electron as a function of laser intensity. Inset: electron kinetic energy (eV) distribution for $I = 5$ MW/cm^2.

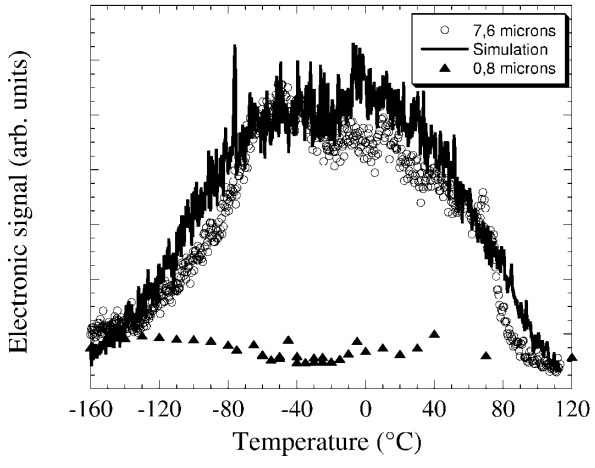

FIG. 2. Electronic signal as a function of the sample temperature (arbitrary units) (•): experiment at 7.6 μm; —: Monte Carlo simulation at 7.6 μm, 5 MW/cm^2, 2 ps pulse duration; (▲): experiment at 800 nm using a Ti:sapphire laser.

using ultrashort (40 fs) Ti:sapphire (800 nm) laser pulses in the GW/cm^2 range. In these conditions, one enters in the multiphotonic regime where there is no reason to expect a dependence of the signal with the temperature, as proven experimentally (Fig. 2). In addition, we have observed an electron current presenting the well known characteristic signatures of the "quantum" regime [9,10]: the photocurrent behaves as the intensity at the power four as four photons are necessary to eject an electron and the maximum current is obtained with p polarized wave. This extra experiment has allowed us to verify that there is no artifact in the setup.

The direct comparison between the experimental result in the midinfrared and the lucky-electron simulation is also addressed in Fig. 2. The parameters are those of our experimental conditions (see Fig. 1) and small shot-to-shot laser intensity fluctuations (less than 10%) were read from the experimental files. The best fit has been obtained with the following expression for the temperature dependence of the MCT: $\tau(\text{fs}) = 8,\ 6 - \frac{T\,^{\circ}\text{C}}{19}$. The comparison between the experiment and the simulation is quite satisfactory. We thus conclude that this simple model is able to provide for temperatures between -160 and 120 °C, a well suited function which was not obvious *a priori*. Note that the choice of a linear dependence is somewhat arbitrary; an exponential function should work as well. We do not claim that we provide a very accurate value of the scattering rates but we must now check that they are reasonable. In the low temperature range (-200 °C), we find a MCT of 20 fs, whereas for the highest temperature (100 °C) we find 3 fs. The first case corresponds to $\omega\tau \sim 2\pi$ and the second to $\omega\tau \sim \pi/4$. The condition $\omega\tau \sim \pi$ occurring when the current is maximum is achieved for $\tau = 12$ fs, the corresponding temperature being ~ -50 °C.

We compare in Fig. 3 our MCT estimates [from the above $\tau(T)$ fit] with what is known in the literature [6,7,11]. Let us observe first that there is a spreading of measurements. Second, we obtain the correct order of magnitude as well as the general temperature behavior. Third, we see that electron-electron collisions (independent of the temperature) do not overwhelm the electron-phonon collisions. Finally, our scattering rates are larger than those measured by other authors. Indeed, they have measured the scattering rates in the static [7] or in a low frequency limit [6,11]. This involves only electrons very close to the Fermi level, and due to the exclusion principle they are not sensitive to electron-electron collisions. In our case, our data should be strongly influenced by electron-electron collision because a transient electron excitation density is generated. Then, as measured explicitly in gold [2], our scattering rate must be larger. Finally note that the 0 K mean free path, known to be sample dependent, is of the order of 40 nm, which is quite reasonable.

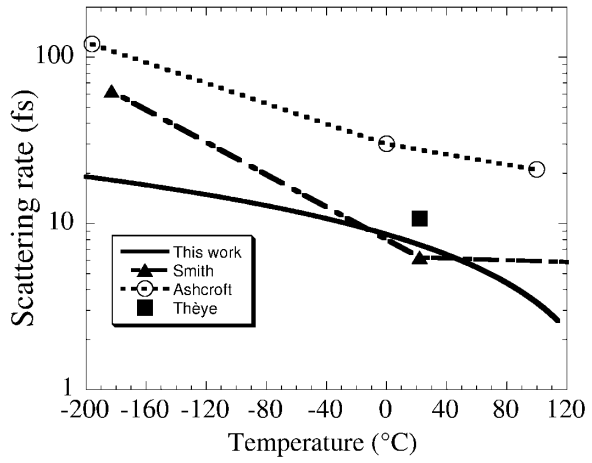

FIG. 3. Mean collision times (fs) for various sample temperatures corresponding to different works.

By using a simple extension of the Drude model, we have been able to extract from a photoemission experiment valuable information concerning the effective scattering rates for the conduction electrons in a pure gold monocrystal in a still unexplored part of the electromagnetic spectrum. The lucky-electron model allows us to explain the temperature dependence of the total electron signal and provides an intuitive description of the energy exchange mechanisms between the electrons and the laser field. Another bonus is that it permits us to derive a single effective collision time with no need for a precise description of the detailed scattering mechanisms. Because the basic physics underlying or description is based on the behavior of the product $\omega\tau$, an interesting issue would be to vary the temperature for different excitation wavelength and thus to observe the shift of the emission peak. This opens new perspectives toward exploring the dynamics of laser-induced electron transport in metals in a still unexplored wavelength region.

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