

## Transport and dynamics of optically excited electrons in metals

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**Abstract.** Time-resolved two-photon photoemission, based on the equal-pulse correlation technique, is used to measure the energy relaxation and the transport of the photoexcited carriers in thin Ag and Au films. The energy-dependent relaxation time shows a significant thickness dependence in the Ag film, whereas for Au a much smaller effect is observed. These experimental observations are compared with a theoretical model based on the Boltzmann equation, which includes secondary (Auger) electrons and transport. A good agreement between experimental and theoretical results is found for Au. However, in our calculations, we did not find any significant change in the thickness dependence in the case of Ag. In order to explain the strong effect in Ag, we discuss the possibility of surface excitations.

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The dissipation processes of photoexcited carriers in metals has been the subject of intense experimental and theoretical work for several decades. The transient behavior of excited electrons at or near metal surfaces is fundamental to many important physical phenomena, including chemical reactions, transport, and molecule–surface interactions, as well as in technological applications of electronic materials. Knowledge of the material and energy dependence of scattering times and, hence, the escape depthlengths of excited electrons is essential for the quantitative interpretation of several surface analytic methods, such as photoemission spectroscopy, Auger electron spectroscopy and low-energy electron diffraction.

In the case of metals whose unoccupied orbitals are not restricted by a band gap, the photoexcited electrons relax directly to a thermal equilibrium with the whole electron gas, randomizing both the energy and momentum of the electrons.

Therefore, the lifetime of individual excited electronic states is always short, typically in the order of only a few femtoseconds to a few tenths of femtoseconds. Consequently, a detailed study of relaxation phenomena in the time domain has just begun [1–4] and many unresolved questions and controversies still exist.

The time-resolved two-photon photoemission technique (TR-2PPE) is the most suitable one for a study of excited-state electron dynamics because it combines energy and momentum sensitivity with a temporal resolution that is only limited by the laser pulse duration [1]. In principle, TR-2PPE determines the depletion of an excited-state population due to inelastic scattering events. There are two effects that affect the measured dynamics: As a direct consequence of the excitation process, secondary electrons will be created by the decay of excited electrons into lower energetic states (cascade electrons) and the refilling of the excited holes (Auger decay electrons). Due to the surface sensitivity of the photoemission technique, the transport of the excited carriers out of the surface region into the bulk (Fig. 1a) appears as an additional decay channel in these measurements [3].

The aim of this publication is to shed light on how the transport of photoexcited electrons out of the surface region influences the experimental data obtained from a TR-2PPE experiment. As transport adds to the system-intrinsic decay due to inelastic scattering, it always reduces the measured lifetime with respect to the actual one. For example, even if the electrons had an infinitely long lifetime, there would still be an experimentally measurable decay in the photoelectron yield as a function of pump-probe delay time due to the electron moving away from the surface and out of the probe volume. If we exclude refilling processes (cascade and Auger electrons) and assume within a first-order approximation separate mechanisms for decay and transport we can add up the measured decay rate in accordance with Matthiessen's rule:

$$\frac{1}{\tau_{\text{meas.}}} = \frac{1}{\tau_{\text{trans.}}} + \frac{1}{\tau_{\text{decay}}} \quad (1)$$

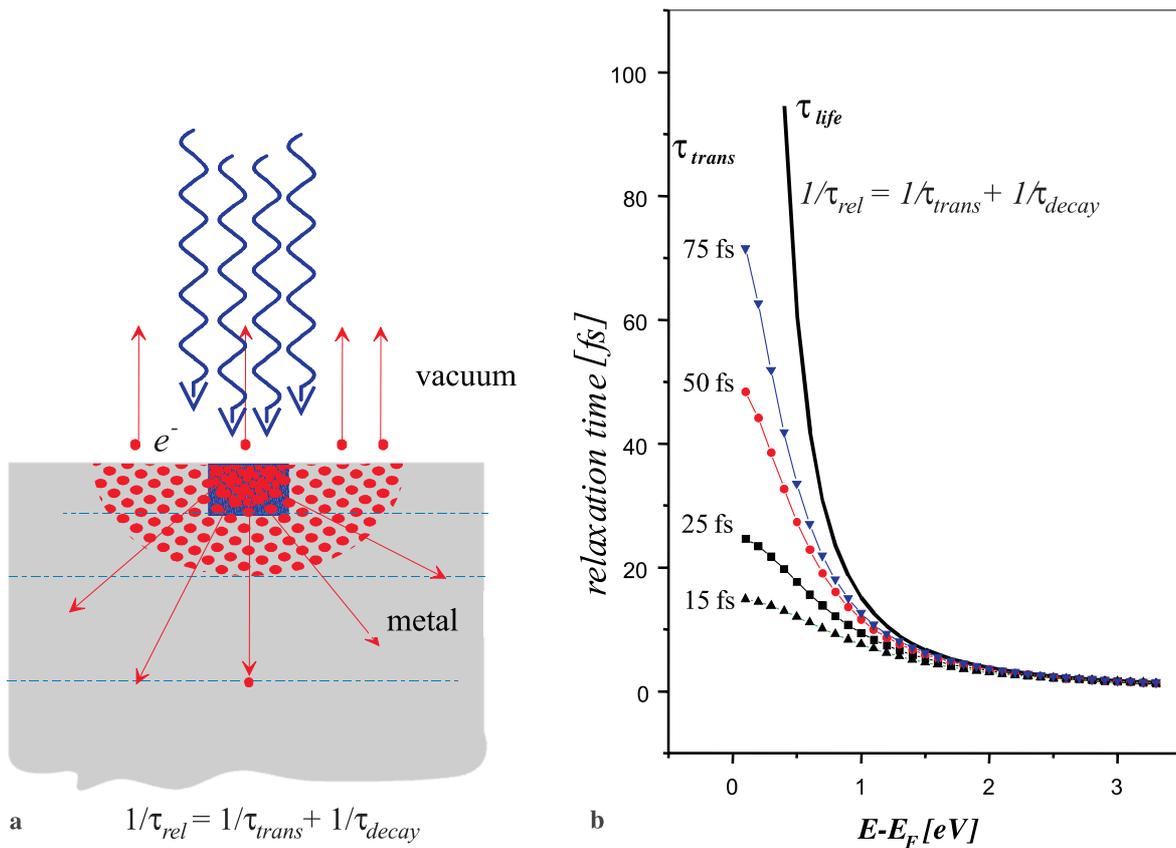
leading to  $\tau(\text{measured}) < \tau(\text{decay})$  as shown in Fig. 1b. Hence, the depletion of an excited-state population in the

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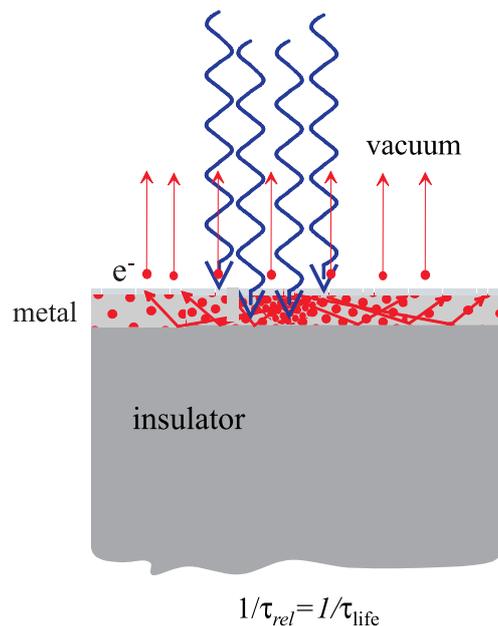
**Fig. 1.** **a** schematic representation of the transport effect in a 2PPE experiment. **b** reduction of the relaxation time caused by the diffusion of electrons into the bulk

probed surface region as a function of the time delay between the two pulses depends on the intrinsic decay of the excited energetic state *and* transport. According to this simple model, it should be possible to reduce or even eliminate the transport effect by using films as thin as the penetration depth of the laser light (see Fig. 2). However, it has to be stressed that this simple rule is only valid provided that the secondary-electron contribution can be neglected [5].

Interestingly, in a recent TR-2PPE study, Cao et al. could not find any thickness dependence in single-crystal Au(111) films at all using film thickness in the range of 15 nm to 300 nm [6]. In their unexpected results they explained that electron transport is a much slower dynamical process in the near-surface region than expected from bulk properties. In contrast to this, however, pump-probe reflectivity measurements on gold films show a significant effect of transport of excited electrons [7, 8]. Hohlfeld et al. showed in a recent publication that their reflectivity measurements could only be explained by a two-temperature model if the effect of ballistic transport is taken into account [8]. They found that a significant carrier redistribution due to transport takes place within the first 100 fs. At least for the lower range of excitation energies (range of longer inelastic lifetime) in the TR-2PPE data of [6] this effect should be visible. Note also that theoretically the effect of transport is still subject to controversy [9].

In this paper, we will compare experimental and theoretical results for TR-2PPE thickness studies of Au and Ag films. We show that the transport of single excited carriers out of the surface region has a significant effect on the relax-

ation time measured by a TR-2PPE experiment. Furthermore, we could not find any indication of a difference between the transport of optically excited electrons in the near-surface region and bulk electrons. We observe, however, a significant



**Fig. 2.** A possibility to reduce or even eliminate the transport effect in a TR-2PPE measurement: using thin films with  $d \sim \lambda_{opt}$

difference in the film thickness dependence of measured relaxation time between silver and gold. A possible explanation might be the large number of secondary (Auger) electrons created in Au under the experimental conditions used. These secondary electrons are not excited in the case of silver because the photon energy used is too low for  $d$ -electron excitation. In order to test this explanation, we compare our data with a theoretical model based on the Boltzmann equation.

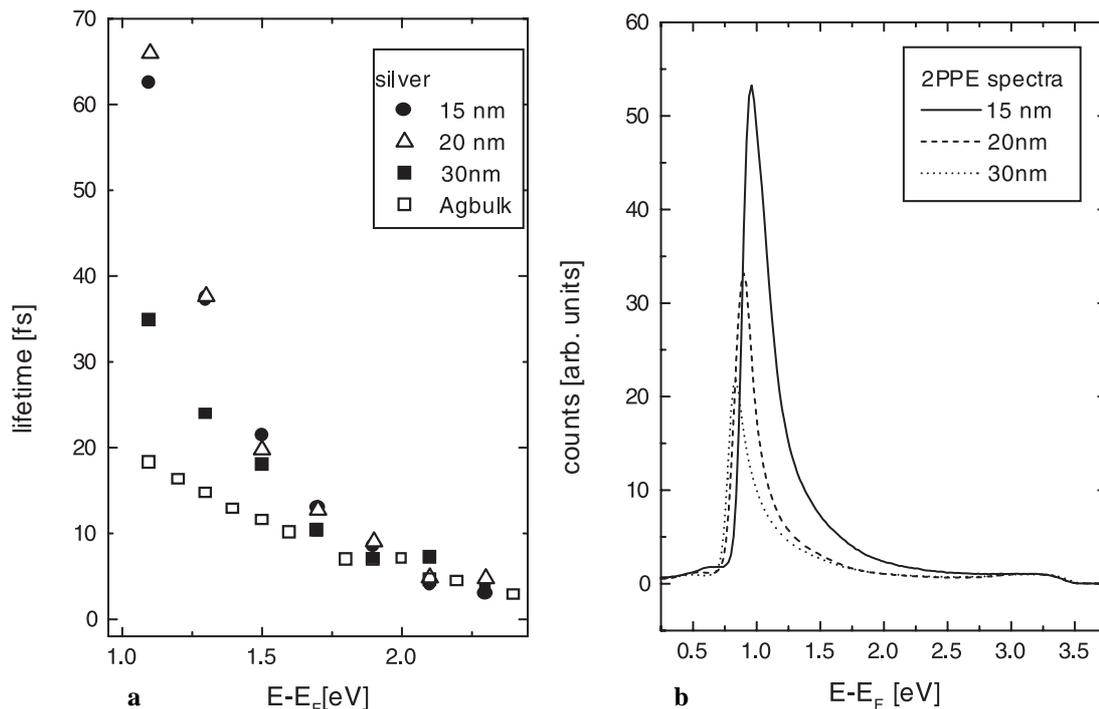
## 1 Experimental setup

The laser system used for our TR-2PPE experiments was a mode-locked Ti:sapphire laser, pumped by about 9 W from a cw Ar<sup>+</sup>-laser. The system delivers transform-limited and  $\text{sech}^2$  temporally shaped pulses of up to 15 nJ/pulse with a pulse width (FWHM) of 40 fs and a repetition rate of 82 MHz. The wavelength can be tuned to a range of 740 to 830 nm. The linearly polarized output of the Ti:sapphire laser is frequency-doubled in a 0.2-mm-thick Beta Barium Borate (BBO) crystal to produce UV pulses of  $h\nu = 3$  eV to 3.4 eV. The UV beam is sent through a pair of fused silica prisms to pre-compensate for pulse broadening due to dispersive elements such as lenses, beam-splitters and the UHV-chamber window in the optical path. The pulses were split by a beam-splitter to equal intensity (pump and probe pulses), and one path could be delayed with respect to the other by a computer-controlled delay stage. The polarization of the pump and probe beams were kept perpendicular to each other ( $s$  and  $p$  polarized) in order to minimize the coherence effect. Both beams were combined colinearly by a second beam-splitter and were focused at 45° with respect to the surface normal of the sample. Photoemitted electrons were detected

along the surface normal ( $k_{\parallel} = 0$ ) by a cylindrical sector energy analyzer.

The data are reconvoluted from the experimentally obtained cross-correlation traces using a rate equation model for the population of the intermediate state. Assuming an exponential depletion of the nascent photoexcited electron population in the energy state  $E$ , the evolution of the transient population  $N(t, E)$  of the intermediate state is given by  $dN(t, E)/dt = A(t, E) - N(t, E)/\tau(E)$  where  $A(t)$  is the excitation induced by the first (pump) laser pulse and  $\tau(E)$  the energy-dependent measured relaxation time. A more correct description of the excitation process within a TR-2PPE experiment requires a quantum-mechanical approach within the density matrix formalism. In the case where the dephasing time of the excitation process is much shorter than the length of the exciting laser pulse this description is equivalent to the above-mentioned rate equation model. At least in the case of bulk electron excitations in metals, as investigated in this publication, and under the used experimental conditions (excitation pulse length= 40 fs), rapid dephasing of the excitation process is a reasonable assumption. All measured cross-correlation curves were symmetrical regarding time zero, which indicates that we really investigate the bulk state (wave packet description, see Sect. 3) rather than the longer-lived surface state. Therefore the impact of transport should not matter whether the probe is  $s$  or  $p$ -polarized.

The epitaxial Ag(100) thin films were grown on air-cleaved MgO(100) single-crystal substrates under ultrahigh vacuum conditions (base pressure  $< 2 \times 10^{-8}$  Pa) and in situ controlled by LEED [10]. The MgO(100) single crystals were transferred into a UHV chamber immediately after cleaving and cleaned by heating to 1300 K for at least 4 hours. The epitaxial Ag(100) films were grown on the substrates at room temperature. The thickness of the film was con-



**Fig. 3.** **a** Measured relaxation times of excited electrons plotted versus the energy above the Fermi level for three different Ag film thicknesses. For comparison, also the bulk value of Ag is shown. **b** 2PPE spectra for the three different film thicknesses

trolled by a quartz-crystal microbalance. This procedure results in smooth, well-defined orientated Ag(100) electrodes with sharp LEED patterns. The Au(111) single-crystal films were fabricated by epitaxial growth on an air-cleaved NaCl crystal.

All films (Au, Ag) were evaporated onto the substrate in a separate chamber. In order to obtain a clean surface, each sample was slightly sputtered and annealed to remove the surface contaminants introduced during transport in the open air. The sample states were checked by Auger and LEED. The bulk samples (Au, Ag) were commercial high purity polycrystalline metal sheets, cleaned by standard sputter and heating cycles.

## 2 Experimental results for thin Ag and Au films

The relaxation of photoexcited electrons as a function of their energies for thin Ag-films with thickness of 15 nm, 20 nm and 30 nm and the data of our bulk sample are shown in Fig. 3a. The photon energy used for these measurements was 3.3 eV. A clear difference in the measured relaxation is visible below  $E - E_F < 1.6$  eV, indicating that in this energy range both the energetic decay of the optically excited electrons and transport reduce the number of detectable photoelectrons. In Fig. 3b, the 2PPE-intensities for all three films are shown. A distinct reduction in intensity with increasing film thickness is observed, caused by a strong diffusion of the excited electrons out of the probed surface region. For the thinner films the potential barrier, set up by the insulating substrate on the reverse side of

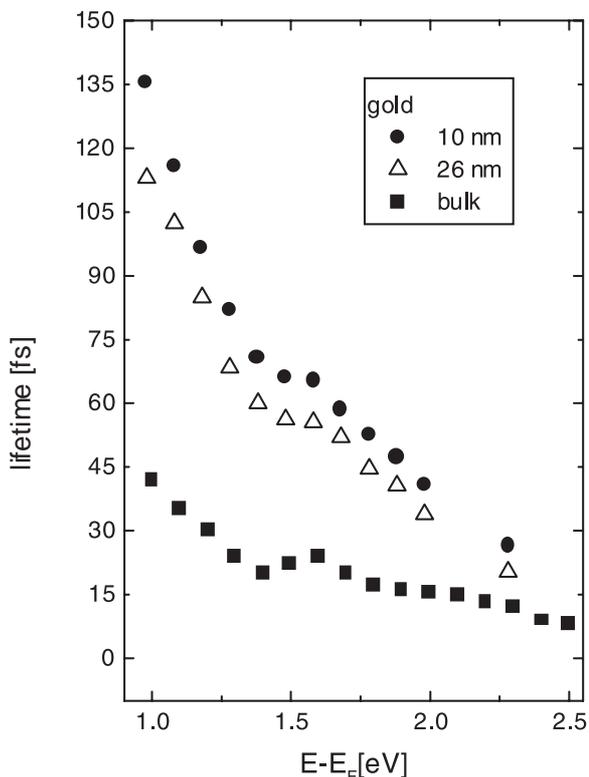


Fig. 4. Measured relaxation time  $\tau$  of excited electrons for various Au film thicknesses and for a Au gold foil

the sample, reduces or even eliminates this diffusion effect and traps the excited electrons within the probed surface area. The intensity change is only apparent for lower excitation energies and hence longer inelastic lifetimes, where the electron mean free path starts to exceed the respective film thickness. Interestingly, we see also an increase in the 2PPE intensity of a 15-nm film compared to a 20-nm film, where we actually cannot find any difference in the measured lifetime.

Figure 4 shows the relaxation measurements of single-crystal Au(111) films for 10 nm and 26 nm, compared with the data of a polycrystalline bulk sample. Two points are noticeable. First, a small change in the slope is visible at about 1.5 eV, which is associated with the photoexcitation of  $d$ -electrons as discussed for copper in [11]. Second, the energy-dependent relaxation for the two thin films shows a significantly smaller thickness dependence compared to the Ag films.

Our experimental data support the intuitive assumption that the transport of excited electrons into the bulk affects the measured lifetime within a TR-2PPE experiment. Depending on the investigated energy region it has either no influence (high excitation energies) on the experimental data or it dominates the measured relaxation dynamics (low excitation energies in the case of silver). Qualitatively, we observe this effect for both of the investigated metal samples, i.e. gold and silver. The quantitative difference in the effect of transport between gold and silver is somewhat puzzling, especially as it appears to be so significant. However, this observation seems to match the observation of an anomalous behavior of the electron lifetime dependence in copper (and gold), reported first by Pawlik et al. [11]. In the light of this discussion, the deviation in the thickness dependence of Au and Ag may be caused by the same effect. The increased relaxation time of Au and Cu compared with Ag can be explained by the fact that for Cu and Au, the electrons in the intermediate state originate predominantly from  $d$ -hole Auger-electrons and not from directly excited photoelectrons out of the  $sp$ -bands (intra-band transitions). How can this affect our transport measurements? The effective mass of the  $d$ -hole is in the order of  $10 m_e$  in some flat regions of the  $d$ -band. This also means that compared to the excited  $sp$ -electrons, the transport of  $d$ -holes away from the surface into the bulk is quite diminished and,

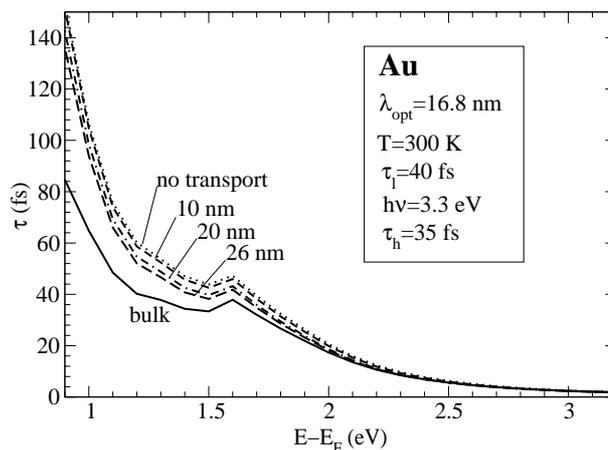


Fig. 5. Calculated relaxation time in Au films of different thickness

consequently, the transport affects the population decay of excited-electrons in the probe-surface region less than it does in the case of excited free carriers. In the following section, we will study the effect of secondary electrons on the transport as determined by a time-resolved 2PPE measurement, by means of a theoretical model based on the Boltzmann equation.

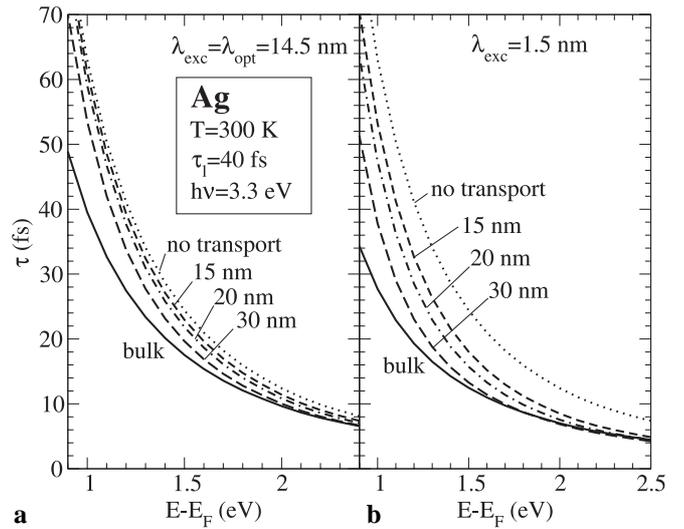
### 3 Numerical Results

The theoretical model we use is based on solving the Boltzmann equation in the random- $k$  approximation which implies that electrons can be described as localized wave packets rather than extended Bloch states (bulk states). This picture is strongly supported by transport measurements [7, 8] which clearly show that electrons are excited close to the surface and propagate into the bulk at approximately the Fermi velocity. In contrast, if electrons were properly described as extended Bloch states, the notion of electronic transport would be meaningless. The random- $k$  approximation also implies that we do not use the energy-momentum relationship for bulk crystals, but an isotropic distribution of momenta for each energy. Consequently, there is no strong distinction in photoemission for single crystals with different surface orientations. This is in agreement with experimental measurements [4]. Furthermore, due to the existence of a surface, the translational invariance is broken (the momentum is not a good quantum number) which invalidates the Bloch-state representation. However, in contrast to assuming Bloch states with definite momentum, in the case where the electronic states contain a superposition of different momenta, one can observe a photoemission current for symmetry directions where the band structure shows a band gap. This is also supported by 2PPE experiments. A further consequence of a wave-packet description of the electrons is that the optical absorption is not momentum specific, but rather isotropic.

Note that there is a difference between one- and two-photon photoemission. In contrast to 2PPE, the measured 1PPE current directly reflects the band structure and  $k$ -resolved measurements are possible. One possible explanation for this difference is that an intermediate level is involved in 2PPE, which seems to make a wave-packet description more appropriate. This is a crucial problem for the theoretical description of 2PPE which merits further study.

The theoretical model includes several relevant processes such as secondary-electron generation, transport, and electron-phonon scattering. The theory was successfully used for the calculation of relaxation times in noble and transition metals [12]. It is able to reproduce and provide an understanding for the existence of the peak in the relaxation time in Cu [13] and can explain its unusual temperature dependence [14]. Thus it is a suitable tool to analyze the effect of film thickness. For the details of the theoretical approach we refer to [12, 14].

In Fig. 5, we consider the case of Au and compare the relaxation time in the bulk and thin films. For the case of films, we assume that particles cannot escape into the substrate through the metal-insulator interface but that they are perfectly reflected at the interface. We consider a lattice temperature of  $T = 300$  K and a laser pulse  $\tau_l = 40$  fs du-



**Fig. 6a,b.** Calculated relaxation time in Ag films of different thickness. **a** Optical excitation within the usual optical penetration depth,  $\lambda_{\text{opt}} = 14.5$  nm. **b** Optical excitation only within  $\lambda_{\text{exc}} = 1.5$  nm to simulate a surface excitation

ration and  $h\nu = 3.3$  eV photon energy, an optical penetration depth of  $\lambda_{\text{opt}} = 16.8$  nm [15] and a Fermi velocity of  $v_F = 1.4$  nm/fs [16] for the  $s$  electrons. For the  $d$  electrons, we use a much smaller velocity,  $v_d/v_s = 0.1$ , in accordance with their large effective mass ( $m_d/m_s = 10$ ). Note that at  $T = 300$  K, the average time between two elastic collisions is  $\tau_{\text{e-ph}} = 1/\Gamma \approx 24$  fs, where  $\Gamma$  is the electron-phonon scattering rate [14].

As a reference, we also show the relaxation time without transport, which corresponds to the case of a uniform excitation of the sample. We can see a small peak at 1.6 eV due to the Auger process, which is discussed in detail in [11–13]. For the thinnest film of thickness  $d = 10$  nm, we observe almost no change in comparison to the case without transport. This is expected, since it takes approximately  $t = 2d/v_F = 14$  fs for an electron to travel back and forth in the film. This gives a lower bound for the time it takes for the density of excited electrons to become uniform across the film. Since it is much shorter than the duration of the pump pulse, the relaxation time will not be affected by transport in such a thin film. Interestingly, for the 20-nm and 26-nm film, the transport is already effective. Here, we find a significant decrease in the relaxation time, which is almost independent of energy. It takes approximately  $t = 2d/v_F = 30$  fs to travel back and forth (of the order of the pulse duration). In addition, the difference in the local density of excited electrons between the front and the back surface of the film is much larger, which will increase the time required to reach a uniform density. Note that for such thin films, the electron motion is purely ballistic, i.e. electron-phonon collisions are irrelevant. Indeed, the latter only start to be effective for thickness  $d \geq v_F \tau_{\text{e-ph}} \approx 36$  nm. For the bulk case, the relaxation time decreases further and the effect is more pronounced at low energy. For example, at 1.5 eV, the relaxation time decreases from 45 fs to 34 fs (22%), whereas at 1 eV it decreases from 110 fs to 64 fs (42%). Note that at 1.5 eV, the variation between the case without transport and the 26-nm film is approximately identical to the one between the 26-nm film and the bulk, which is in agreement with the

experimental results. Notice that in the bulk case, the electron motion is diffusive for sufficiently low-energy electrons, i.e. for relaxation time  $\tau \geq \tau_{e-ph}$  [14]. Otherwise, the electrons have a short lifetime and do not feel the presence of phonons.

So far we have considered the case of Au, where electrons from the  $d$ -bands are excited ( $d$ -band threshold at  $-2$  eV). To analyze the role of the  $d$ -holes, it is interesting to analyze the case where  $d$ -bands are not involved. Ag is the perfect candidate since its  $d$ -band threshold lies at approximately  $-4$  eV below the Fermi surface. In Fig. 6a, we have plotted the relaxation time for bulk and thin films for an optical excitation with  $\lambda_{opt} = 14.5$  nm. The peak found for Au is absent in the case of Ag, due to the fact that no transitions from the  $d$ -band are possible for photon energy  $h\nu = 3.3$  eV. Otherwise, the results for Ag and Au are very similar. In particular, the magnitude of the variation in film thickness is almost identical for both materials. This strongly indicates that the film-thickness dependence of the relaxation time is not influenced by the excitation of  $d$ -bands. In other words, with respect to transport, it does not matter whether the excited electrons are predominantly Auger-type or primary. In Fig. 6b, we show results for an optical excitation only within  $\lambda_{exc} = 1.5$  nm to simulate a surface excitation, which is discussed below.

#### 4 Comparison between experimental and theoretical results

The experimental results show that the effect of transport in Au thin films is small but significant in contrast to the strong reduction observed in Ag. We have found a reasonably good agreement between experimental and theoretical results in the case of Au. However, regarding the thickness dependence, our calculations did not show any difference between Au and Ag. Let us make some comments and discuss possible scenarios to explain the strong difference between Au and Ag observed experimentally.

##### 4.1 Effects of $d$ -bands

The presence of Auger electrons in Au, but not in Ag, could have been the origin of a discrepancy between Au and Ag. However, our calculations, which are able to describe both situations, have clearly excluded this possibility. On the contrary, we have found that the thickness dependence of the relaxation time does not depend on whether the excited electrons are Auger-type or primary.

##### 4.2 Influence of secondary electrons

Although the  $d$ -bands are not excited in Ag, our calculations have shown that other secondary (cascade) electrons are still predominant. It is not entirely clear what is the contribution of secondary electrons to the excited electrons. It is interesting to see whether in the case where no secondary electrons are involved, the film-thickness dependence of the relaxation time is strongly modified. In order to test this possibility, we have calculated the relaxation time without secondary-electron generation. The results show that for films with  $d < 30$  nm, the transport effect is almost unchanged.

##### 4.3 Effects of elastic scattering and film quality

It is possible that the film quality might be responsible for the difference between Ag and Au. If we assume that the Ag samples are of lower quality than the Au films, this can be simulated in our model by increasing the elastic scattering rate. This is formally equivalent to increasing the temperature of the lattice. We have seen that including elastic scattering, for example electron-phonon scattering at  $T = 300$  K, strongly reduces the effect of transport [14]. Note that for films with  $d < \lambda_{opt}$ , the samples are excited almost uniformly from the beginning and the transport is absent. Only for thicker samples, will the elastic scattering rate play a role and increase the relaxation time. Thus, within this picture one cannot understand the strong thickness dependence observed in thin Ag films.

##### 4.4 Effect of the optical penetration depth

All these observations show that the main reason for the insensitivity to transport in the case of thin films is the large optical penetration depth ( $\lambda_{opt} = 15$  nm). However, we have seen that for the bulk, the effect of transport and elastic scattering can lead to very strong effects on the relaxation time. Hence, the important parameter that determines the strength of the transport effect is the ratio between the excitation depth  $\lambda_{exc}$  of electrons and film thickness. Thus if  $\lambda_{exc}$  is much smaller than the film thickness, we expect bulk behavior. The smaller this ratio, the more significant is the transport effect even at small film thickness. In order to illustrate this point, we have analyzed the relaxation time in Ag using a very small excitation depth of  $\lambda_{exc} = 1.5$  nm (see Fig. 6b). We have found a much stronger transport effect for thin films ( $d < 30$  nm) than in the case of  $\lambda_{opt} = \lambda_{exc} = 15$  nm, as shown in Fig. 6b [5]. Therefore, to explain the pronounced transport effect in silver at low film thickness (10, 30 nm) in comparison with gold, we have to find a mechanism that makes the excitation depth for silver much smaller than for gold. A consequence of the position of the  $d$ -bands in silver (inter-band contribution to the dielectric function) is an anomalous, low-energetic surface-plasmon resonance of about 3.8 eV in the case of smooth surfaces [15, 17, 18]. That is why the excitation of the surface-plasmon resonance with the 3.3-eV energy photons is a possible mechanism for creating a very surface-located distribution of excited electrons. Surface plasmons are known to decay mainly by interaction with the electron gas into single electron excitations [19]. This mechanism would obviously lead to a much more surface-located electron excitation and consequently reduce the excitation depth/film thickness ratio. However, in the case of gold this mechanism is extremely improbable for the used excitation energy of 3.3 eV. For gold only a free-electron plasmon resonance can be found. For noble metals this resonance is located at about 8 eV excitation energy [17, 20].

Note that surface-plasmon excitation is strongly enhanced on rough surfaces, where translational invariance parallel to the surface is broken and momentum conservation is no longer required [21]. Already the slight sputtering of the smooth sample ( $< 1$  nm) prior to measurement might have been enough to introduce a significant amount of plasmon creation on our sample and thereby reduce the excitation

depth. However, for a deeper understanding further work regarding the dynamics of plasmon excitations on noble-metal surfaces is required.

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