

is claimed in ref. 1. Angular-resolved ellipsometry measurements on the sample would help to clarify the scenarios that are applicable here.

Furthermore, the presence of artefacts in the Au layer would affect the electron mobility and have direct consequences for the modelling of a superdiffusive spin current. This goes beyond the scope of our Correspondence.

In conclusion, we have given a general approach for calculating the absorption profile of multilayered thin films. In particular, this approach can disentangle thermal and non-thermal phenomena in complex multilayer structures. By applying this approach, we demonstrated

that in ref. 1 mistakes were made in the calculation of the light absorption in a Au/Ni/Pt/Al layered structure, which has led to a strong underestimation of the light absorption in the Ni layer. Therefore, the conclusion that “in striking contrast to existing knowledge, a direct optical excitation is not a precondition for ultrafast demagnetization” cannot be substantiated by the presented experiment. \square

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Eschenlohr *et al.* reply — We welcome the Correspondence by Khorsand *et al.*¹ as it gives us the opportunity to correct a mistake in our Article² and clarify further our findings. The main criticism raised by Khorsand *et al.* concerns the estimated pump-laser absorption in individual layers of our Au/Ni sample.

In our Article² we used time-resolved X-ray magnetic circular dichroism experiments to probe the femtosecond demagnetization in metallic multilayers after laser irradiation. The samples we investigated consisted of a 15-nm Ni film capped with 30-nm Au, and an uncapped 20-nm Ni film used as

reference. We estimated the amount of laser light reaching the Au and Ni layers using the Beer–Lambert law (in differential form) and the IMD software³: $I(z) = |E(z)|^2 = I_0 e^{-\alpha z}$ (with absorption coefficient α , depth z , intensity transmitted through the surface I_0 , and electric field E), which suggested that in the Au-capped sample only 7% of the absorbed power reaches the Ni layer.

However the relevant quantity for the excitation of the layers is given by the variation in the real part of the Poynting vector⁴, resulting in the transmitted power $P(z) = n(z)|E(z)|^2 = n(z)I_0 e^{-\alpha z}$, with n the real part of the complex index of refraction $\tilde{n} = n + ik$. In our original calculation the coefficient $n(z)$ was missing, but we agree with Khorsand *et al.* that it is needed in order to correctly calculate the absorbed laser power in multilayered samples. Before examining the consequences, we emphasize that our experimental results are independent of the calculated absorption profile and hence do not need correction.

With the tabulated optical constants of pure Au ($n = 0.18$, $k = 4.91$, at 780-nm wavelength)⁵, the amount of absorbed power is 49% in Au, 37% in Ni, and 14% in the Pt buffer and Al substrate (Fig. 1a). For clarity we normalize these numbers to the total absorption of the sample, as in our Article². Notably, this does not neglect the intrinsic reflectance R of the sample surface. Calculating R for the Au/Ni sample with the tabulated values results in 0.95 (35° incidence, p-polarization)³. However, the measured R is 0.88 (ref. 2), that is, our sputtered gold film is not perfect. The origin of the reduced reflectivity can be the polycrystalline microstructure and/or defects, which have

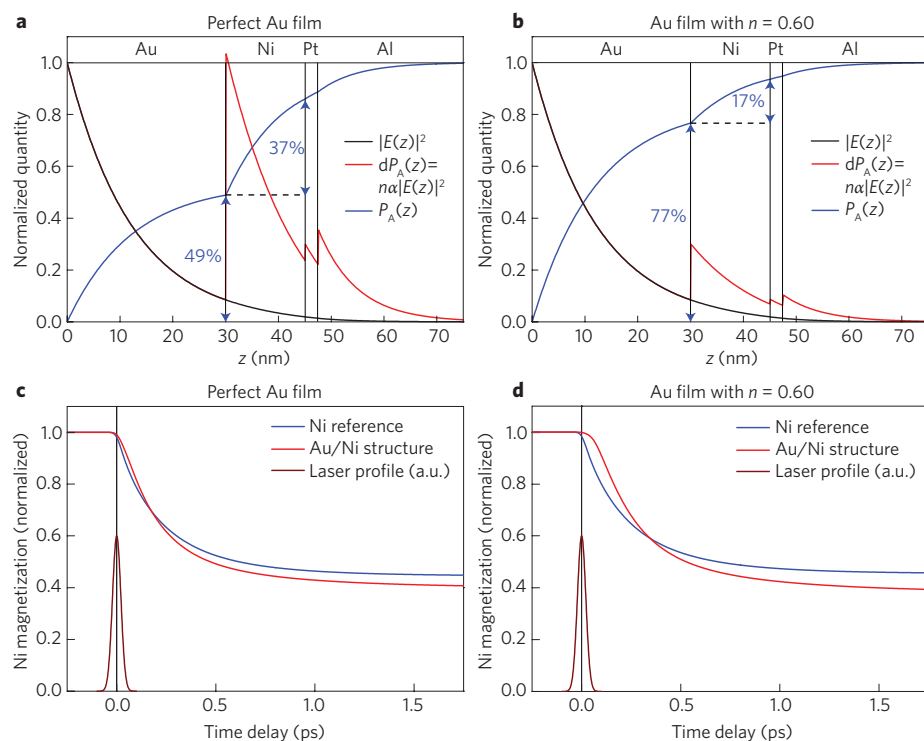


Figure 1 | Calculated cross sections and time responses. **a,b**, Field intensity $|E(z)|^2$, differential absorbed power $dP_A(z)$ and absorbed power $P_A(z)$, normalized to 1 at $z = 0$ (E , dP_A) or $z = \infty$ (P_A) for a perfect Au film (**a**) and for an imperfect Au film with $n = 0.60$ (**b**). The percentage of absorption in individual layers is marked by the arrows. **c,d**, Time evolution of the average magnetization of the Ni layers in the Au/Ni structure and the Ni reference sample for the excitation scenarios shown in **a,b**. Calculation for a perfect Au film (**c**) and for an imperfect Au layer with $n = 0.60$ (**d**). Both were computed for absorbed fluences of 3.9 mJ cm^{-2} for the Ni reference and 1.6 mJ cm^{-2} for the Au/Ni structure, respectively.

been shown to cause a higher absorption⁶ for photon energies of 1–2 eV. Accordingly, a demonstrated way⁶ to describe the imperfect Au film is by a modified dielectric constant $\epsilon = \tilde{n}^2 = n^2 - k^2 + 2ink$. As $\text{Re}(\epsilon) = \epsilon_1 = n^2 - k^2$ changes very little⁶, and $n \ll k$, we can keep $k = 4.91$ fixed and find, for $R = 0.88$ and using IMD (ref. 3), a value of $n = 0.60$. This results in 77% absorbed power in Au and 17% in Ni (Fig. 1b), not dramatically different from our original numbers of 90% and 7% for Au and Ni, respectively². In our view, the four different scenarios given by Khorsand *et al.* to explain the reduced R remain hypothetical, whereas the scenario we propose has been proven experimentally for Au (ref. 6).

Both absorption profiles are used as input for the superdiffusive transport calculation. For perfect Au, shown in Fig. 1c, fits give time constants of 230 fs for Ni, 235 fs for Au/Ni and 10 fs delayed response of Au/Ni with respect to the Ni reference. The calculations used a corrected simulation code, which slightly changes the time for Ni compared with the 250 fs in our original Article. The fitted time constants for imperfect Au, shown in Fig. 1d, are 230 fs for Ni, 260 fs

for Au/Ni and a 50 fs delay. The latter hot-electron-transport calculations agree very well with the experimentally observed slower demagnetization and delayed response of the Au/Ni sample. Conversely, the calculation for perfectly crystalline Au only poorly describes the experimental result. Especially the higher laser excitation of Ni leads to a considerable amount of immediate demagnetization, in contrast to the experimental results that show a clear delay. We believe that our model of superdiffusive hot-electron transport applied to a non-perfect Au film with modified optical constants is the only possible explanation of the observed delay in the magnetic response in Au/Ni, as well as its reduced reflectivity. Any other proposed mechanism of ultrafast demagnetization would not result in the experimentally observed time delay. In particular this underlines our original conclusion that demagnetization can be accomplished by fast transport of non-equilibrium hot electrons. By focusing only on the absorption calculation and ignoring experimental results, the comment by Khorsand *et al.*¹ falls short of disproving our above arguments. □

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On the thermodynamics of light trapping in solar cells

To the Editor — The important issue of thermodynamic losses in high-efficiency solar cells has been discussed in ref. 1 using an equation (equation (1) in Box 1) that describes how the open-circuit voltage V_{oc} is reduced from its radiative limit.

The first term yields the reference value for the radiative limit under full concentration. The second term contains entropic losses due to radiative emission in directions others than those defined by the solid angle of the Sun. The third term deals with light trapping and the fourth is related to non-radiative recombination where the internal luminescence quantum efficiency $QE = R_{rad}/(R_{rad} + R_{nrad})$ as defined in ref. 1.

We will show in the following that the four terms in equation (1) are not independent of each other and result from contradictory approximations, in such a way that equation (1) cannot be correct. We

start with the well-established reciprocity relation $qV_{oc} = qV_{oc}^{rad} + kT \ln(QE^{LED})$ (refs 2–4) where V_{oc}^{rad} is the V_{oc} value determined by the recombination current that leads to the emission of photons. The quantity $QE^{LED} = J_0^{em}/(J_0^{em} + J_0^{nrad})$ is the external

quantum efficiency of the device when operated as a light-emitting diode (LED), where J_0^{em} and J_0^{nrad} are the saturation current densities caused by photon emission and non-radiative processes (including photon emission followed by parasitic absorption),

Box 1 | Upper limit for the V_{oc} of a solar cell and three entropic loss terms.

$$qV_{oc} \approx E_g(1 - T/T_{Sun}) - kT \ln(\Omega_{emit}/\Omega_{Sun}) - kT \ln(4n^2/I) + kT \ln(QE) \quad (1)$$

$$qV_{oc} = kT \ln \left(\frac{\int A \phi_{Sun} dE}{\int A \phi_{bb} dE} \times \frac{\epsilon_{in}}{\epsilon_{out}} \times \frac{\epsilon_{out} \int A \phi_{bb} dE}{S_{cell} w [R_{nrad} + (1 - p_r) R_{rad}]} \right) \\ = kT \ln \left(\frac{\int A \phi_{Sun} dE}{\int A \phi_{bb} dE} \times \frac{\epsilon_{in}}{\epsilon_{out}} \times \frac{\epsilon_{out} \int A \phi_{bb} dE}{S_{cell} w 4n^2 \pi \int A \phi_{bb} dE} \times \frac{QE}{1 - QE + (1 - p_r) QE} \right) \quad (2)$$

q , elementary charge; E_g , band gap energy; k , Boltzmann's constant; T and T_{Sun} , temperature of the cell and the Sun, respectively; Ω_{emit} and Ω_{Sun} , the spherical angle of the light emitted and absorbed by the solar cell; n , the refractive index; I , concentration factor.