A Comparative Study of Two-Photon-Photoemission Sources

In this appendix we will discuss the perspectives for two-photon photoemission experiments using laser and synchrotron radiation in a pump-probe experimental setup. Formulas for the excitation probability and the total photoemission signal as well as the expected 1-photon background are derived and several conditions and setups are discussed.

A.1 Excitation Probabilities

Firstly, formulas for the excitation probabilities as a function of the number of photons per laser pulse, as well as the reflectivity of the surface, the absorption length and the focus diameter, will be derived. Secondly, several experimental conditions, including laser-laser experiments as well as laser-SR experiments for several laser systems and SR repetition rates, will be compared.

Assuming an amplified laser system with approximatively 1 W power at the fundamental wavelength of 800 nm with a repetition rate of 200 KHz (Seedlaser+Coherent RegA) the number of photons per pulse is about $2 \cdot 10^{13}$ at the fundamental and with a conversion efficiency of 50% about $5 \cdot 10^{12}$ at the 2nd harmonic. For further use, we note that with a typical conversion efficiency of less than 10% we may obtain $5 \cdot 10^{11}$ photons per pulse in the 3rd harmonic.

These laser photons will now be focussed on a surface area A_{sf} given by the laser focus l_{focus} as $A_{\mathrm{sf}} = l_{\mathrm{focus}}^2 = (0.1\ mm)^2$, which corresponds to $6 \cdot 10^{10}$ unit cells in the case of silicon with a unit cell length l_{uc} of 4 Å . The number of photons per surface unit cell n_{uc} is then given as:

$$n_{\mathrm{uc}} = N_{\mathrm{pu}} \cdot \frac{l_{\mathrm{uc}}^2}{l_{\mathrm{focus}}^2} \tag{A.1}$$

where N_{pu} is the number of pump photons per pulse. For the outlined above experimental parameters we obtain:

$$n_{\mathrm{uc}} = 10^{13} \cdot \frac{(4 \cdot 10^{-10})^2}{(100 \cdot 10^{-6})^2} = 1.6 \cdot 10^2 \frac{\text{photons}}{\text{surface unit cell}} \tag{A.2}$$

These photons are either reflected by the surface or absorbed by the solid-state sample. Introducing the reflectivity $\mathcal{R}$ and the absorption length l_{abs}, the absorption will be equivalent to the excitation probability P_{exc}:

$$P_{\mathrm{exc}} = (1-\mathcal{R})\frac{l_{\mathrm{uc}}}{l_{\mathrm{abs}}} \cdot n_{\mathrm{uc}} = (1-\mathcal{R})\frac{l_{\mathrm{uc}}}{l_{\mathrm{abs}}}\frac{l_{\mathrm{uc}}^2}{l_{\mathrm{focus}}^2} \cdot N_{\mathrm{pu}} = (1-\mathcal{R}) \cdot \frac{V_{\mathrm{uc}}}{V_{\mathrm{abs}}} \cdot N_{\mathrm{pu}}\,. \quad \text{(A.3)}$$

where V_{uc} and V_{abs} are the volumes of the unit cell and the absorption region, respectively. For $\mathcal{R} = 90\%$, $l_{\mathrm{abs}} = 1\ \mu m$, $l_{\mathrm{uc}} = 4$ Å, $l_{\mathrm{focus}} = 100\ \mu m$ and $N_{\mathrm{pu}} = 10^{13}$ an excitation probability of 0.6% has been obtained. Assuming that no further complications due to selection rules or short life times aggravate the experiment, excitation of this order of magnitude should be visible in a photoemission experiment.

Typical count rates are about 1000 counts/s for filled states under single bunch conditions (with the laser on every 6'th single bunch pulse and using gating electronics for the suppression of the unpumped signal). Therefore one would expect a countrate about 6 count/s for excited states.

A.2 Background Signal

Another aspect is the problem of the background signal. Background occurs from several sources: Dark counts from the channeltrons, higher harmonics of the synchrotron radiation and the finite energy resolution of the electron analyzer. Dark counts are typically less than 0.1/s and the 2nd harmonic signal from the synchrotron can be suppressed by, e.g, an aluminium filter (and setting the SR photon energy shortly below the Magnesium absorption edge at 35 eV) to less than 10^{-4} of the signal of the filled states in the valence band. The analyzer is operated typically at a resolution of 200 meV (10 eV pass energy and 5 mm slits). This means that the signal above the Fermi level is suppressed by approximatively $\exp\left(-(\Delta E_{HL}/\Delta E_A)^2\right)$, where ΔE_{HL} is the homo-lumo energy difference and ΔE_A is the electron analyzer resolution. At $\Delta E_{HL} = 0.5$ eV this factor is $e^{-6.26} \approx 2 \cdot 10^{-3}$ at $\Delta E_{HL} = 1$ eV this factor is 10^{-11}. This illustrates, that the energy spread of the analyzer is an important source of one-photon background as long as the excited state is close to the Fermi edge.

For the approximation in table A.1, the following formula for the background count rate r_{bg} has been used:

$$r_{\mathrm{bg}} = c \cdot R_{\mathrm{pr}} N_{\mathrm{pr}} \cdot 10^{-4}\,. \quad \text{(A.4)}$$

where, R_{pr} is the repetition rate and N_{pr} is the number of photons of the probe pulses. The factor 10^{-4} describes the ratio between the signal from a filled state to the background above the Fermi level. The constant c reflects the photoemission efficiency as well as the analyzer transmission. To estimate this constant, we compare

the filled state photoemission-count-rate r_{pe} for conditions we have in single-bunch mode with the analyzer electronics gated with 208 KHz. There $N_{\mathrm{pr}} = 10^6$, r_{pe} is typically 1000 counts/s and therefore:

$$c = \frac{r_{\mathrm{pe}}}{r_{\mathrm{photon}}} = \frac{r_{\mathrm{pe}}}{N_{\mathrm{pr}} R_{\mathrm{pr}}} = \frac{1000}{10^6 \cdot 208 \cdot 10^3} = 5 \cdot 10^{-9} \tag{A.5}$$

This constant can be thought of as the overall photon efficiency of the photoemission experiment including the electron analyzer.

A.3 Two-Photon Signal

The formula for the two-photon photoemission signal are easy derived:

$$r_{\mathrm{2PPE}} = c \cdot P_{\mathrm{exc}} \cdot R_{\mathrm{pu}} N_{\mathrm{pr}} \,, \tag{A.6}$$

which is just the excitation probability times the signal from a filled state. R_{pu} is the repetition rate of the laser pump-pulses. In this formula we assumed, that $R_{\mathrm{pu}} \leq R_{\mathrm{pr}}$ and every pump pulse matches one probe pulse.This is true for the RegA system, where the laser pulses match every 6th single bunch but not for the Ti:Sa oscillator running at 83 MHz in single bunch, where only every 200th pump pulse matches one probe pulse. Collecting terms, the most important parametric dependencies are (from the point of view of the experimental design):

$$r_{\mathrm{2PPE}} \propto R_{pu} \cdot \frac{N_{\mathrm{pu}} N_{\mathrm{pr}}}{l^2_{\mathrm{focus}}} \tag{A.7}$$

This formula explicitly shows the linear dependence of the two-photon signal on the pump as well as the probe intensity. Laser-laser experiments for example reach much higher probe intensities than a SR-based experiment. Additionally, 1-photon background can be completely suppressed by choosing the probe photon energy below the work function of the sample. Assuming that the average power is conserved in an amplified laser system, it has to be noted on passing, that the 2PPE-signal is inversely proportional to the repetition rate, because N_{pu} and N_{pr} are each proportional to R^{-1}_{pu}. This is not true for the laser-SR experiment, because there the probe intensity can not be increased above the single bunch value of 1.25 MHz. Further amplification for example by the RegA system will lead to higher excitation probabilities, but the total two-photon signal will not increase. Only the one-photon background can be reduced.

A.4 Comparison of Different Setups

In table A.1 we compare several experiments. Two laser-laser setups are considered, an unamplified system with 75 MHz repetition rate and an amplified system with

Setup	N_{pu}	N_{pr}	l_{focus}/m	P_{exc}	r_{2PPE}/s^{-1}	r_{bg}/s^{-1}
lr-lr, unamp.	10^{11}	10^{9}	$5 \cdot 10^{-5}$	$2.5 \cdot 10^{-4}$	10^{5}	0
lr-lr, amp.	10^{13}	10^{11}	$5 \cdot 10^{-5}$	$2.5 \cdot 10^{-2}$	$3 \cdot 10^{6}$	0
Ti:Sa @ U125-SGM, SB	$2 \cdot 10^{9}$	10^{6}	10^{-3}	$1.3 \cdot 10^{-8}$	$3 \cdot 10^{-5}$	0.62
Vanadat @ U125-SGM	10^{12}	10^{6}	10^{-3}	$6.4 \cdot 10^{-6}$	$4 \cdot 10^{-2}$	0.62
Vanadat @ U125-PGM	10^{12}	10^{6}	10^{-4}	$6.4 \cdot 10^{-4}$	4	0.62
RegA @ U125-PGM,not gated	10^{13}	10^{6}	10^{-4}	$6.4 \cdot 10^{-3}$	6.6	0.62
RegA @ U125-PGM, gated	10^{13}	10^{6}	10^{-4}	$6.4 \cdot 10^{-3}$	6.6	0.10
Ti:Sa @ U125-PGM, MB	10^{11}	10^{4}	10^{-4}	$6.4 \cdot 10^{-5}$	1.0	12.5

Table A.1: Comparison of several experimental conditions for 2PPE experiments.

250 KHz repetition rate, respectively. The focus in both cases is assumed to be 50μm. The laser-SR setups include the Vanadat laser with 1.25 MHz repetition rate in single bunch under the old conditions at the U125-SGM (1 mm Focus) and the actually conditions at U125 PGM beamline (0.1 mm focus). The numbers for a cavity dumped Ti:Sa with 1.25 MHz repetition rate and 80 μJ pulse power would of course be very similar. Additionally the RegA system with 208 KHz repetition rate with and without gated analyzer electronics as well as an unamplified Ti:Sa oscillator synchronized to every 6th bunch in multi bunch ($R_{\mathrm{pu}} = 63$ MHz, $R_{\mathrm{pr}} = 500$ MHz) are taken into account. The estimated photon numbers per pulse (N_{pu} and N_{pr}) and focus diameters are found in the table as well as the calculated excitation probabilities, the two-photon signals r_{2PPE}, and the background r_{bg}. All excitation probabilities are calculated for a 4 Å unit cell, a surface reflectivity of 90% and an absorption length of 1 μm using the Equations A.3, A.4 and A.6.

A.5 Conclusion

Several aspects can be discussed after this comparison. One of the main motivations was the question, why laser-laser experiments are so successful. The reason is, as seen in the table that even so these experiments also work with low excitation probabilities, they get much more two-photon signal simply due to the several orders of magnitude higher number of probe photons. Additionally, their advantage is the complete absence of one-photon background. The price they pay for this is certainly the very small accessible binding energy and momentum range.

Another aspect for the design of laser-SR experiments is, that further amplification of the laser for the price of lower repetition rate will not increase the 2PPE-signal, even so gating electronics can reduce the background and therefore increase the signal to noise ratio. Without gating the Coherent RegA system has almost no advantage in comparison to the Vanadat or a comparable cavity dumped Ti:Sa system running at 1.25 MHz. The higher excitation density is compensated by the

lower repetition rate. Even an unamplified Ti:Sa oscillator would deliver comparable two-photon signal in multi-bunch mode, but the expected one-photon background would make experiments difficult. In this case gating out the right probes pulses is not so easy and the multi-probe setup has other disadvantages to be discussed elsewhere.

Nevertheless, the calculations show that with the RegA system in single bunch we should have a fair chance to see a few counts of two-photon signal on almost zero background. The condition for this is a stable laser system, an adjustable focus of less than 0.1 mm and last but not least a system with appropriate absorption parameters and lifetimes. For the assumed absorption length of 1 μm and reflectivity of 90% we expect an inversion of about 1%. Higher absorption and the absence of surface reflectivity might increase this number.

B RKKY-Interaction

The RKKY-interaction stands for Ruderman-Kittel-Kasuya-Yosida and plays a role in the magnetic spin ordering of gadolinium, with its strong localized magnetic moment of the 4f ion cores and the lack of angular momentum (S=7/2, L=0), classifying gadolinium as a Heisenberg ferromagnet. The exchange interaction between electrons can be expressed by the Heisenberg operator.

$$\widehat{H} = -\sum_{ij} J(R_{ij}) S_i \cdot S_j \tag{B.1}$$

where J is the exchange integral related to the overlap of the charge distribution of the localized atoms. The minus sign favors the parallel (ferromagnetic) orientation. The localized 4f electrons (which form the permanent magnetic moments) cannot interact via direct exchange coupling due to the extremely small overlap between the wave functions of the 4f electrons. The 4f electrons can more effectively interact via indirect exchange coupling through the valence band polarizing the electrons in valence band. This is the RKKY-interaction. Mathematically, the coupling from the delocalized s-electrons (spin σ_j) with the f-electrons (spin S_j) at the same position j is described by the s-f model (Kondo-lattice model):

$$\widehat{H} = -J \sum_{j} S_j \cdot \sigma_j \tag{B.2}$$

The induced exchange splitting of the ($5d^1 6s^2$) valence-band states is proportional to the 4f magnetization. $\widehat{H}$ collapses for temperature reaching the Curie temperature. A complete calculation of the dependence of the gadolinium-band structure on temperature was done by Rex *et al.* [Rex99].

B.1 Conservation Rules

The hamiltonian has to conserve the total angular moment of the system:

$$\mathbf{J} = \mathbf{L}_e + \mathbf{S}_e + \mathbf{L}_p + \mathbf{L}_w \tag{B.3}$$

where $\mathbf{L_e}$ is the angular momentum of the electrons, $\mathbf{S_e}$ is the spin-angular moment of the electrons, $\mathbf{L_p}$ is the angular momentum of the phonons and $\mathbf{L_w}$ is the angular

momentum of the photons. The conservation of angular momentum is one of the mysteries of instantaneous laser-induced demagnetization. The spin and orbital moment of the electronic system are related to its magnetic moment. The total magnetic moment μ is:

$$\boldsymbol{\mu} = \boldsymbol{\mu_B}(\mathbf{L_e} + \boldsymbol{g}\mathbf{S_e}) \tag{B.4}$$

with the Landé g-factor ($g = 2$). Since the total Hamiltonian of the system conserves the total angular momentum, a change in magnetization can only be achieved by exchange among the four contributions on the right-hand side of Equation B.3. A classical experiment by de Haas and Einstein in 1913 [Ein15] demonstrated that the induced magnetization will be compensated by a rotation of the body, i.e., an exchange occurs between S and L_p. Moreover, spin-orbit (SO) coupling is necessary. Without its presence there is no spin-lattice relaxation that converges the precessing motion of the electron spins towards a net magnetization parallel to the applied field. Deducing from this experiment, a transfer of the spin of the electronic system after laser illumination to the lattice angular moment would allow to explain fast demagnetization conserving the total angular momentum **J** [Koo03].

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