A Spin analysis

One of the crucial points in data processing is a correct spin analysis. Parameters, which have to be assigned prior to the experiments are the spin sensitivity of the detector given by the Sherman function S and in some cases the detector-related asymmetry in the count rates R (see Section 3.1.2). After having demonstrated the importance of a correct spin analysis with a showcase example we will discuss in the following sections several methods to deduce S and R.

A.1 Sherman function and detector related asymmetry

The spin polarization can be extracted from the count rate asymmetry in the channeltrons N_1 and N_2 of the SPLEED detector by

$$P = \frac{1}{S}\frac{N_1 - N_2}{N_1 + N_2} \tag{A.1}$$

(see Section 3.1.2). An erroneous determination of the Sherman function S does not only lead to a systematic error in the spin polarization, but also influences the measured exchange splitting and linewidth. This is illustrated in Figure A.1 for a spin-resolved measurement of 3 ML Fe/Cu(100) which was evaluated with three different Sherman functions. The exchange splitting deduced from the spectrum with $S = 0.3$ rises from 54 meV to 77 meV for an extreme choice of $S = 0.19$.

A different efficiency of the counters would also have a serious impact on the data analysis and lead, e.g., to an apparent spin polarization of an unpolarized electron beam. In spin analysis this problem can be circumvented by two ways:

- Recording two spectra with antiparallel spin polarization usually provided by opposite magnetization direction of the sample. The detector related asymmetry is canceled by taking the geometric mean value of the count rates in both measurements (see Section 3.1.2). This, however, does not hold for the p-polarized measurements for cobalt films due to dichroism.
- Determination of the detector-related asymmetry defined here as $R = N_1^0/N_2^0$ and normalizing the count rates according to this asymmetry. With this method a possible erroneous assignment of the initial asymmetry contributes additionally to the overall systematic error in spin polarization.

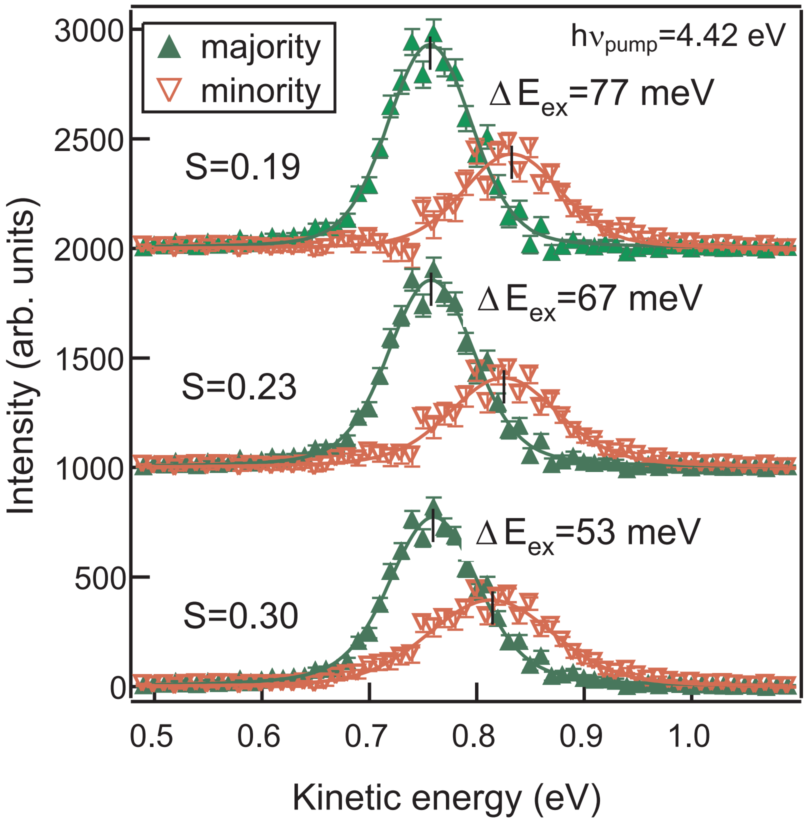

Figure A.1: *Influence of the Sherman function shown for a spin-resolved 2PPE measurement of a 3 ML iron film on Cu(100). The spectra are obtained from the same raw data and only differ in the choice of the Sherman function in data processing.*

A.2 Determination of the Sherman function

A.2.1 Highly spin-polarized electron beam

In this work it was only possible to estimate a lower limit of $S = 0.23$ whereas an upper limit of $S = 0.33$ is given by theoretical calculations [*Kirschner and Feder*, 1979]. The lower limit could be deduced from measurements with a highly spin-polarized beam which was provided by s-polarized measurements of cobalt films. Using a Sherman function smaller than $S = 0.23$ in data evaluation resulted in an unphysical spin polarization of $P > 1$ for a specific region in the spectrum. This can also be observed in Figure A.1 for $S = 0.19$. At the low energy side of the minority peak a negative count rate is evident and therefore $S = 0.19$ can definitely be ruled out.

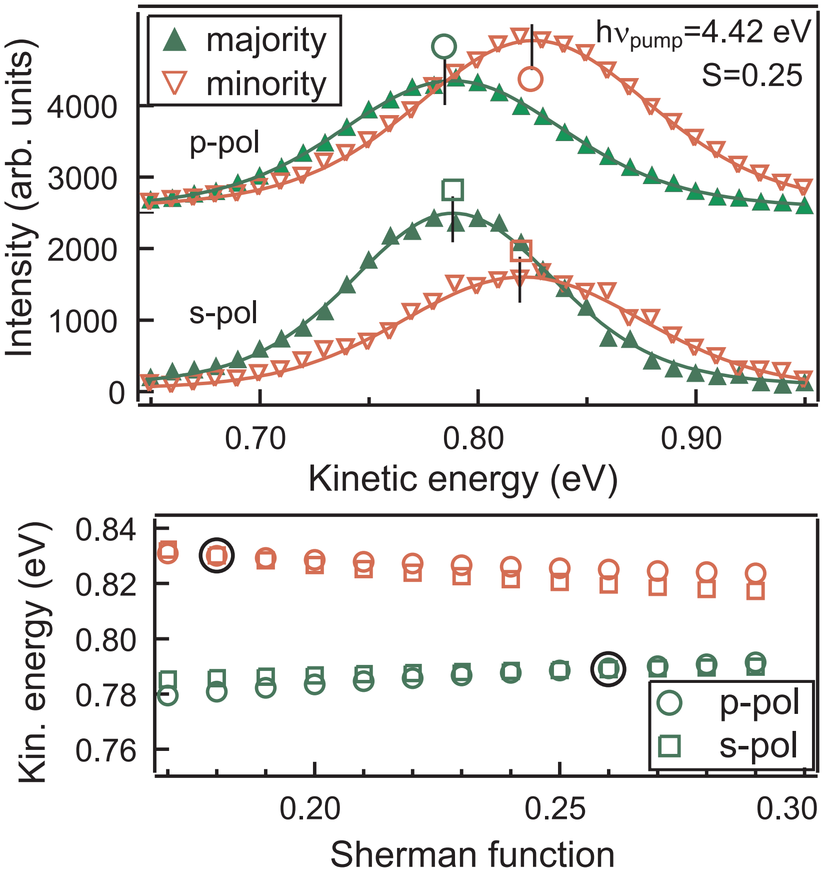

Figure A.2: *Determination of Sherman function with p- and s-polarized measurements. Upper panel: Spin-resolved spectra of 3 ML Fe/Cu(100) for p- and s-polarized pump-light obtained with a Sherman function of* $S = 0.25$. *The pins mark the peak maximum. Lower panel: Peak maximum versus the Sherman function. The black circle denote an equivalent position for the majority and minority peak maximum, respectively.*

A.2.2 Recording spectra with different light polarizations

One possibility to deduce the Sherman function is given by a comparison of the energetic peak position of the n=1 image-potential state between p- and s-polarized measurements. As shown above, the determined majority and minority peak positions depend on the choice of the Sherman function. This dependence is, however, different for spectra taken with p- and s-polarized light, because of the different spin polarization in the n=1 image-potential state (see Figure A.2). Equal positions must be obtained for a correct choice of the Sherman function. However, with respect to the two spin components an ambiguous result can be seen. While the energetic positions of the minority peaks are equal for a Sherman function of $S = 0.18$ this is only true for the majority peaks deduced with $S = 0.26$. The failure of this method lies in the assumption that the peak position is independent on the light polarization. For still overlapping pump- and probe pulses the electronic structure of the initial states influences the peak position in 2PPE, due to excitation via virtual states, especially enhanced close to a resonant excitation (see Section 5.1). This has to be considered for an inhomogeneous distribution of the initial-state band structure, as it is the case for the minority states in iron thin films (see Section 4.2). The influence of the initial states on the peak position in spin-resolved 2PPE is discussed in detail by *Schmidt* [2007]. From the results in Section 4.2 the majority initial states should be distributed more homogenously. Therefore a Sherman function of S=0.26 seems to be more realistic than S=0.18, which yields unphysical features anyway, as discussed in the preceding section. Reliable results would only be obtained for a large pump-probe pulse delay, which is, however, not feasible for s-polarized measurements due to very small count rates.

In data processing we chose a Sherman function of S=0.23 or S=0.24 for the determination of the binding energy in Section 5.1, which is close to the estimated minimum value. This pessimistic estimation considers the fact that the crystal is expected to be slightly degraded. Furthermore this value corresponds to the Sherman function which is obtained by a more elaborate method presented in Section A.3.4

A.3 Determination of the detector related asymmetry

A.3.1 Unpolarized electron beam

To deduce the asymmetry the most straight forward method is given by a calibration with an unpolarized electron beam, which is e.g. provided by 2PPE from Cu(111). A direct comparison between the two channeltron count rates, sensitive to *in-plane* spin polarization, is shown in Figure A.3. The two peaks in the spectra arise from the n=1 and n=2 state. The latter is resonantly excited by the Shockley surface state due to the high photon energy ($h\nu_{pump}$= 4.89 eV). The asymmetry, which we define here just as the ratio of the count rates obtained with an unpolarized beam $R = N_1^0/N_2^0$, can be extracted from the quotient of the integrated count rates. For kinetic energies larger than 0.5 eV the energy dependence of R is small, a deviation in R between the n=1 and n=2 states is smaller than 2.5%. For small kinetic energies the detector related asymmetry however differs and has therefore to be determined separately. This is of special importance for data analysis of electrons in the low energy cut-off.

A critical influence on the asymmetry R has the electron beam path. Therefore care has to be taken that the reference measurements is recorded under equal conditions. For example, placing the coil below the sample influences the elec-

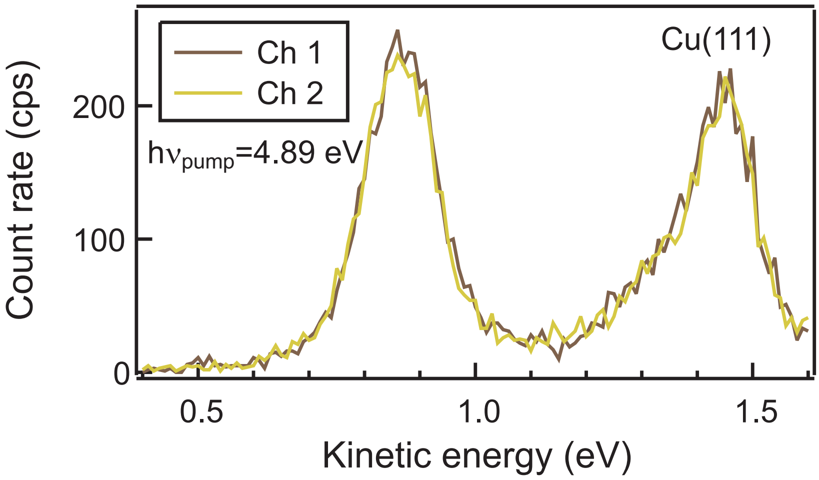

Figure A.3: *Determination of the initial asymmetry with a 2PPE spectrum of Cu(111). Only the two relevant channeltron count rates sensitive to the in plane spin polarization are shown. The low count rate arises from the low pump-pulse power due to the extraordinary high photon energy* $h\nu_{pump} = 4.89$ *eV*

trostatic potential between sample and analyzer, hence the asymmetry changes. The relative intensity is also dependent on the laser spot position with respect to the entrance lens of the analyzer, which needs therefore to be fixed.

A.3.2 Measurement with s-polarized light

As explained in section 4.3, dichroism of *in-plane* magnetized films is only present in spectra recorded with p-polarized light in the present set-up. Only little dichroism is observed with s-polarized light, which is ascribed to a small persisting perpendicular component $A_\perp$ of the vector potential of the pump pulse. Therefore, by changing the magnetization direction also the spin polarization simply reverses its sign. Consequently, the respective channeltron count rates can be directly compared: The count rate in channeltron 1 of the measurement with the magnetization vector pointing upwards (↑) should differ from the count rate of channeltron 2 in the measurement with opposite magnetization direction (↓) by a multiplication factor given by the detector related asymmetry R. The same holds for the other pair of channeltron count rates. The equivalence of the respective s-polarized spectra is illustrated in Figure A.4.

A.3.3 Non-dichroic region of spectrum

Although dichroism usually governs p-polarized spectra, there might be an energetic region with no dichroism. In this case one can proceed in analogy to

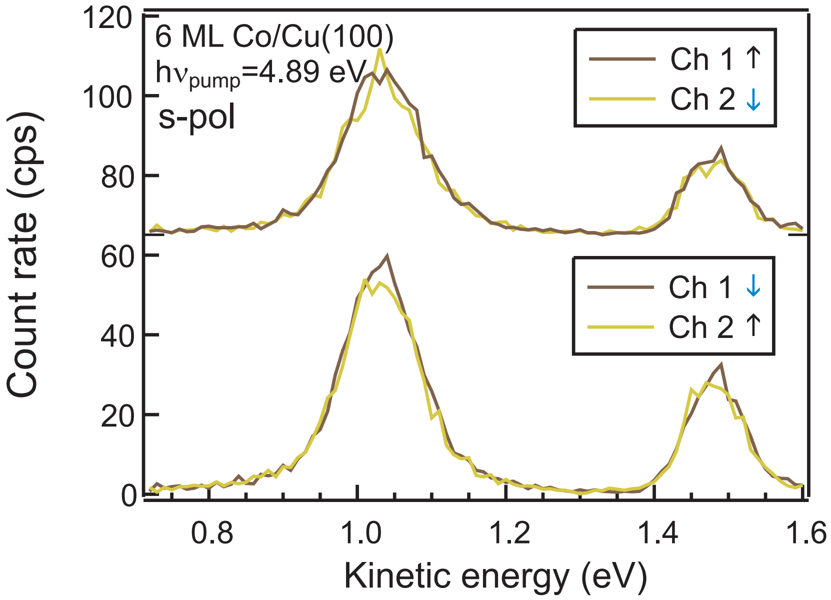

Figure A.4: *Determination of the initial asymmetry with s-polarized measurements on 6 ML Co/Cu(100). Count rates of the opposing channeltrons (Ch1, Ch2), which are measured in opposite magnetization directions (↑↓) can be compared.*

measurements recorded with s-polarized light. This is demonstrated for a p-polarized measurement of a cobalt film recorded with a high pump-pulse photon energy in Figure A.5. In the low energy side of the n=1 state only little dichroism is present (not explicitly shown) and therefore only this energy region can be taken into consideration.

These three methods used to estimate the asymmetry R yield results which are equal within an error of roughly 5%. The deviations may be explained with a still existing dichroism in the s-polarized spectrum or in the specific energy range of the p-polarized spectrum. The determination of R with an unpolarized electron beam provided by photoelectron spectroscopy from the Cu(111) sample may lack an exactly equivalent measurement geometry compared to the Co/Cu(100) sample. For data evaluation in this work R was either taken from measurements on Cu(111) or from s-polarized measurements with no serious impact on the results.

A.3.4 Minority state effect

A nice alternative method to assign the asymmetry R and the Sherman function S simultaneously can be thought of by a dichroic time resolved measurement[1]. As discussed in section 4.3 mainly minority states are believed to be the origin

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[1] Energy-resolved measurements would work as well, but data acquisition time has to be considerably increased and the energy range to be carefully chosen.

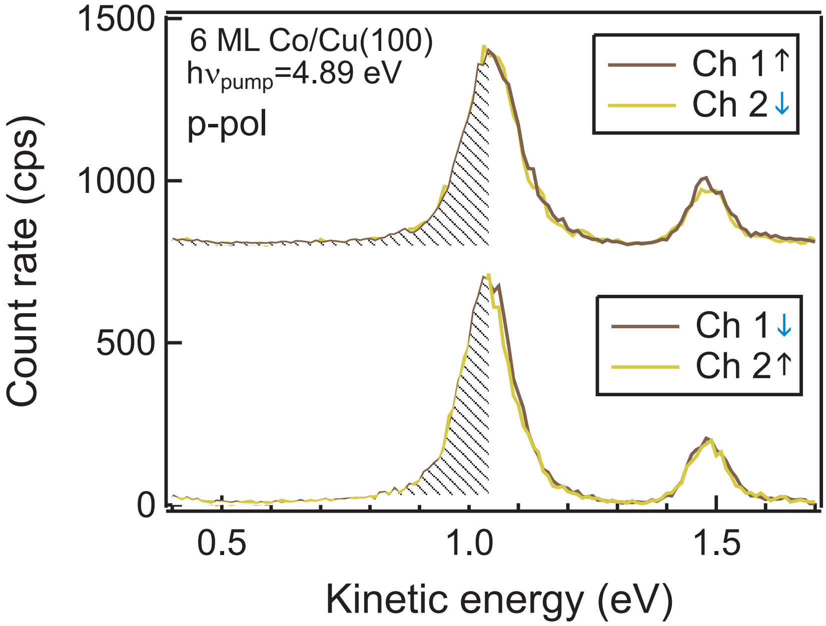

Figure A.5: *Determination of the initial asymmetry with p-polarized measurements on 6 ML Co/Cu(100). Count rates of the opposing channeltrons (Ch1, Ch2), which are measured in opposite magnetization directions (↑↓) can be compared if no dichroism is present (shaded area).*

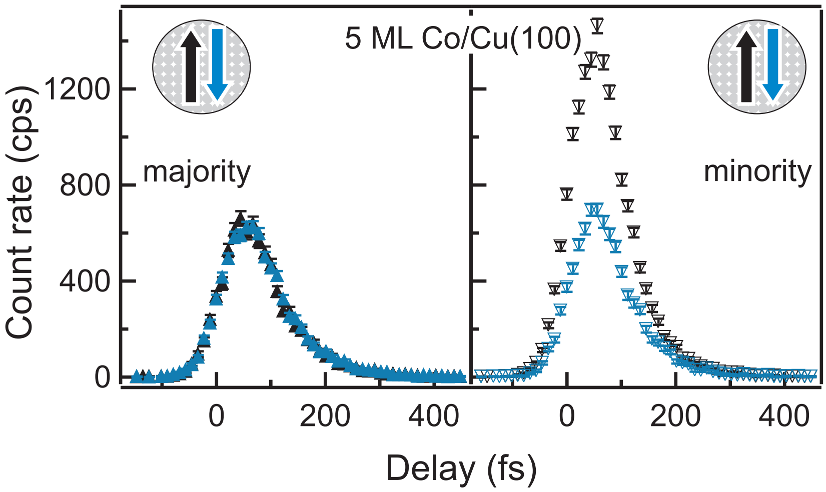

Figure A.6: *Determination of the Sherman function S and initial asymmetry R with time-resolved dichroic measurements on 5 ML Co/Cu(100). S and R have been chosen to obtain identical majority count rates for both magnetization directions. The corresponding integrated measurement is shown in Figure 4.18.*

of the dichroic features. If one now assumes this effect to be caused solely by minority states, one can seek for a Sherman function S and an asymmetry coefficient R which provide the same majority count rate from the data set for both magnetization directions. This is shown for a time-resolved measurement of a 5 ML cobalt film on copper in Figure A.6. With a choice of a Sherman function of $S = 0.24$ and an asymmetry of $R = 0.98$ equal majority count rates are obtained over the whole spectrum. The value for R corresponds within the error to an estimated asymmetry of $R = 1.01$, obtained from measurements on Cu(111). The assumption of a pure minority effect needs a theoretical verification, because any experimental result bases on a well known Sherman function and detector related asymmetry.

Furthermore, this assumption would make it possible to extract the minority lifetime without any spin-sensitive detection. The difference in the count rates of spin-integrated measurements in both magnetization directions should be proportional to the minority count rate. Roughly speaking, a spin sensitivity is already given by the dichroic photoexcitation itself, which only affects minority electrons in this experiment.

B Error analysis

The spectral distribution is subject to various sources of errors, which can be classified in systematic and statistical ones. The most prominent systematic error in this experiment is a possible erroneous assignment of the Sherman function S, as discussed in the previous chapter. This error is not included in the error bar of the data points in the spectra, but considered for the values given for the binding energy and the exchange splitting. This source of error is also not taken into account for the spin polarization which is shown in Figure 4.8 and 4.13. A wrongly determined Sherman function S_{wrong} generally leads to a spin polarization which differs from the true spin polarization by the factor S/S_{wrong} and therefore can be easily corrected afterwards.

The error bar in the spectra only includes the statistical error. This error bases on the detection of individual events (counts), which distribution is described by Poisson statistics. Since the number of counts is always large in the spectral range (typically 50 - 10000 counts) this distribution can be well described by a Gaussian curve with a standard deviation σ which is given by

$$\sigma_i = \sqrt{N_i}, \tag{B.1}$$

where N_i denotes the number of counts in a channeltron. This standard deviation is taken as the error of the counts of the respective channeltrons

$$\Delta N_i = \sigma_i. \tag{B.2}$$

In the case of ultrathin iron films spectra were recorded for both magnetization directions (measurement A and B) and a geometric mean value of N_{1A} and N_{2B} and vice versa was used to account for experimental artifacts as described in Section 3.1.2. In this case the error ΔN_i in Equation B.2 has to be replaced by

$$\Delta N_i = \frac{1}{2}\sqrt{\frac{N_{iA}}{N_{jB}}(\Delta N_{jB})^2 + \frac{N_{jB}}{N_{iA}}(\Delta N_{iA})^2} \tag{B.3}$$

The error of the spin polarization and the spin-dependent intensities can be calculated by error propagation. For the spin polarization P, which is given by the number of counts in the respective channeltrons N_1 and N_2

$$P = \frac{1}{S}\frac{N_1 - N_2}{N_1 + N_2}. \tag{B.4}$$

the error can be deduced to

$$\Delta P = \frac{1}{S}\sqrt{\left(\frac{2N_2\Delta N_1}{(N_1+N_2)^2}\right)^2 + \left(\frac{2N_1\Delta N_2}{(N_1+N_2)^2}\right)^2} \tag{B.5}$$

.

The spin-dependent intensities ($I_{\uparrow\downarrow}$) are given by

$$I_\uparrow = \frac{1+P}{2}\left(N_1+N_2\right) \tag{B.6}$$

$$I_\downarrow = \frac{1-P}{2}\left(N_1+N_2\right). \tag{B.7}$$

By error propagation one obtains

$$\Delta I_\uparrow = \sqrt{\left(\frac{S+1}{2S}\Delta N_1\right)^2 + \left(\frac{S-1}{2S}\Delta N_2\right)^2} \tag{B.8}$$

$$\Delta I_\downarrow = \sqrt{\left(\frac{S+1}{2S}\Delta N_2\right)^2 + \left(\frac{S-1}{2S}\Delta N_1\right)^2}. \tag{B.9}$$

From this equation one immediately notices that for $S \rightarrow 1$, i.e. an ideal spin-polarization detector, the error converges to the deviation of the respective counts in the channeltrons, whereas for the opposite case ($S \rightarrow 0$) the error diverges. Therefore, a large Sherman function is a prerequisite for spin-sensitive experiments.