Time-resolved polarization decoherence in metal hole arrays with correlated photons

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We study the combined polarization decoherence experienced by entangled photons due to time- and space-related dephasing processes in a metal hole array. These processes are implemented when the entangled photons are sent through a birefringent delay and are focused on the array. In particular, we demonstrate that compensating the temporal separation of the two polarizations after passage through the array can only partly recover the original coherence. This, surprisingly, shows a coupling between the temporal and spatial decoherence channels; we ascribe this coupling to transverse propagation of surface plasmons. © 2006 Optical Society of America

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1. INTRODUCTION

The use of entangled photon pairs has proved to be a powerful tool for several forms of information processing, such as quantum cryptography, but also for precise optical measurements in the field of quantum metrology. The benefits of these techniques over their classical counterparts is based on the exploitation of the robust correlations, which are often well known, between the photons. In this paper, we report on the use of entangled photons for measurements on surface plasmons in metal hole arrays.

Metal hole arrays (metal films perforated with a periodic array of subwavelength holes) can exhibit extraordinary transmission of light with a certain resonant wavelength. This transmission is surface-plasmon mediated; freely propagating light incident on one side of the metal film excites surface plasmons (SPs) that subsequently couple through the hole pattern to SPs at the other side and finally radiate into photons. Since its original demonstration, this phenomenon has been studied in different contexts, including the survival of quantum entanglement in the mentioned conversion process.

The polarization properties of the extraordinary light transmission in metal hole arrays have been a special topic of study in several papers. Polarization- and angle-resolved measurements in Ref. 12 have shown that propagating SPs can act as polarization selectors, i.e., only transmission of the polarization component that is aligned with this direction occurs. This strong relation between SP propagation and the polarization properties of the extraordinary transmission is also demonstrated in a study of the polarization decoherence as a function of the numerical aperture of the light beam that is focused on the array. This polarization decoherence, which also depends on the incident state of polarization, is ascribed to the propagation of SPs in combination with the non-plane-wave character of the incident beam. The study of surface-plasmon propagation via polarization properties requires an analysis that is most clearly described in terms of four Stokes parameters and a 4×4 Mueller matrix, representing a black-box description of the complicated physical system.

In this paper, we will go beyond the space-resolved polarization decoherence studied in Ref. 13. More specifically, we additionally impose a time delay between the H- and V-polarization components before focusing on the hole array. As we measure the polarization correlations in the 45° basis, this temporal distinction leads to a lower-polarization fringe visibility and thus to an additional polarization-decoherence channel in time (on top of the decoherence in space induced by SP propagation and focused illumination). Our key question in this respect is whether both decoherence channels are independent of each other, i.e., whether, in the presence of focused illumination, the decoherence that is due to the time delay can be fully compensated for by retiming the H and V components behind the hole array.

Within the context of the Mueller-matrix black-box method the answer should be positive, i.e., operations in time and space should be independent of each other and can thus be interchanged at will. However, optical decoherence is different from most other forms of decoherence in the sense that the polarization information is not lost to some abstract (infinitely large) environment but is instead spread over the time and space coordinates within the optical beam; the unpolarized component appears only after temporal and/or spatial averaging over the full beam. As the “labeling” information remains available within the propagating beam, it can be extracted and used in a later stage. As a result, consecutive decoherence channels are not necessarily independent, and the corresponding (4×4) matrices generally cannot be multiplied in a simple way. We will show experimentally that the decoherence channels are indeed coupled in our black box as they are mixed by propagating surface plasmons.

As a final remark, our discussion of the observed decoherence in metal hole arrays is qualitative in nature. The reason for this is that a sufficiently complete and simple
Lee et al.

theory of light transmission through hole arrays does not yet exist. Several (independent) numerical models that would allow a more quantitative analysis are around, but using these would lead to model-dependent results. Instead, we have chosen to focus on the generic features.

2. EXPERIMENTAL METHODS

Figure 1 shows the experimental setup that provides for the generation of polarization-entangled photon pairs via the process of spontaneous parametric downconversion (SPDC). The figure caption describes in detail how our SPDC source generates a polarization-entangled signal-and-idler beam and how the time and space information in one of the beams can be modified by a polarization-dependent (birefringent) delay and focusing onto a metal hole array. For sufficient time resolution in the experiment, it is essential that the spectral bandwidth of the entangled photons is larger than the spectral width of the transmission resonance of the hole array. We have chosen relatively thin (0.25 mm) crystal in order to generate entangled photons over a large spectral bandwidth; with the properly scaled geometry such a thin crystal can generate even more entangled photons than a thicker crystal.

We use a metal hole array in which the holes are arranged in a hexagonal lattice. The hole diameter is 200 nm, and the lattice constant is 886 nm. The holes have been etched with a focused ion beam into a 200 nm thick film of gold that is bonded to a glass substrate with a 2 mm thick titanium layer. The hole array is positioned in the focus between two lenses that form a 1:1 telescope. In the experiment we use only different sets of these lenses behind the apertures to vary the numerical aperture of the light incident on the array. Thereby, the aperture diameter is fixed at 5 mm, giving a detection angle of 25 mrad, which is smaller than the SPDC ring crossings (30 mrad).

A typical transmission spectrum of the hole array at plane-wave illumination is shown in Fig. 2. The very sharp resonance peak of the hole array is essential; its FWHM of only 18 nm is much smaller than the 50 nm spectral width of the interference filter (also shown), which in turn is somewhat smaller than the FWHM of the SPDC light. Please note that in the literature FWHM values of at least 50 nm are reported for (1,1) SP resonances in square arrays. We think that the resonance in our sample is so sharp because we use a hexagonal array instead of a square array; the reciprocal lattice of the hexagonal array is rotated with respect to its direct lattice, which leads to less SP scattering at the holes. Furthermore, our sharp resonance is carried by the SP mode at the air–metal interface, which experiences less damping than that at the glass–metal interface.

To create a time delay \( \tau \) between two orthogonal polarization components of the SPDC light, a set of quartz waveplates and a Soleil–Babinet compensator are placed in the beam. The waveplates are oriented in the directions of the BBO axes and have thicknesses that differ by factors of 2 and range from 0.31 to 4.94 mm (\( \approx 2^4 \times 0.31 \) mm), corresponding to a time delay \( \tau \) range from about 9 to 145 fs. Using polarization interferometry, we have measured the exact thicknesses of the waveplates; these agree very well with the specified values (error in \( \tau < 0.1 \) fs).

The polarization decoherence induced by a temporal separation of the \( H \) and \( V \) components can be characterized by the fringe visibility of the coincidence rate scanned as a function of the time delay \( \tau \). In a typical measurement, we determine the envelope of this fringe pattern, which is defined by minimal and maximal coincidence counts. These minima and maxima are measured when we fine tune \( \tau \) via the Soleil–Babinet compensator (2 \( \pi \) range) such that the optical-path difference between the \( H \)- and \( V \)-component is precisely \( N \lambda \) and \( (N+1/2) \lambda \), respectively, since our compensated SPDC source is set to the singlet two-photon state \( |HV> - |VH>\rangle/\sqrt{2} \). Here, \( \lambda \)
is the degenerate wavelength of the SPDC light, being 2 × 407 = 814 nm. We measure with the fast axes of the waveplates in both horizontal (negative τ) and vertical (positive τ) directions. From the measured coincidence counts \( R_m \) at path difference \( m\lambda \), we calculate the polarization fringe visibilities \( V \) via \( (R_{m+1/2}−R_{m})/(R_{m+1/2}+R_{m}) \). We note that negative time-resolved visibilities can be obtained (see Fig. 3), as sidelobes of a sinc profile, owing to the rectangular shape of the spectrum of the interference filter (see Fig. 2). Whenever possible, we scanned the Soleil–Babinet compensator over more than \( \lambda/2 \) to find the exact minima and maxima. The error in the measured visibilities \( V \) is typically 0.01 and is caused by quantum fluctuations in \( R_N \) and \( R_{N+1/2} \).

3. EXPERIMENTAL RESULTS

Figure 3 depicts the time-resolved visibility measurements, performed with the time delay in front of the telescope (see Fig. 1). We first concentrate on the solid curve in Fig. 3 which shows the measurement without the hole array. This curve has a peak visibility of \( V = 0.96\pm 0.01 \), which quantifies the entanglement quality produced with our SPDC source.\(^{19} \) The high visibility shows that complications due to entanglement in transverse momentum are avoided, as the apertures used in the experiments are smaller than the size of the SPDC ring crossings. The visibility decays sharply with the time delay \( \tau \) the small width of 65±2 fs (peak to zero) is associated with the spatial domain due to the polarization-dependent propagation of surface plasmons out of the limited region excited with a focused optical beam.\(^{13,20} \) Equivalently, this reduction can be ascribed to the combined polarization and angle dependences of the optical transmission.\(^{12} \) The fact that we observe lower visibilities with increasing time delay \( \tau \) shows the additional polarization decoherence in the time domain. The decrease is sharpest for the case of strongest focusing (NA=0.15), where we obtain a peak-to-zero width of 76±2 fs. For the cases NA=0.053 and NA=0.017, the low-viscosity values decay much more gradually, and the approximate zeros are less accurate. Therefore we instead determine the peak-to zero width as 88±2 fs and 160±8 fs, respectively, for these cases.

In Fig. 4 the averaged absolute values of \( V(−\tau) \) and \( V(+\tau) \) shown in Fig. 3 are plotted on a logarithmic vertical scale as a function of \( |\tau| \). The thicker curve without markers represents the measurement without hole array. The straight solid line is a fit of the exponentially decaying part of the NA=0.017 curve, from which a decay time \( \tau_c = 38\pm 1 \) fs is obtained.

result from the sharp edges in the "top hat" transmission spectrum of the interference filter (see Fig. 2).

Next, we positioned the hole array in the center of the telescope (see Fig. 1). The three marked curves in Fig. 3 show time-resolved visibility measurements with a hole array for three different numerical apertures (NA) of the light incident on the hole array. First, we see that the peak visibility \( V \) drops from \( V = 0.93\pm 0.01 \) (NA=0.017) by \( V = 0.83\pm 0.01 \) (NA=0.053) to \( V = 0.73\pm 0.01 \) (NA=0.15). We ascribe this reduction to polarization decoherence in the spatial domain due to the polarization-dependent propagation of surface plasmons out of the limited region excited with a focused optical beam.\(^{13,20} \) Equivalently, this reduction can be ascribed to the combined polarization and angle dependences of the optical transmission.\(^{12} \) The decay of the NA=0.017 curve is described very well by a simple exponential \( a \exp(−\tau/\tau_c) \) with a decay time of \( \tau_c = 38\pm 1 \) fs. For this case of weak focusing, the measured decay time is just the field decay time of the surface plasmons. At a propagation speed of \( \approx 0.95c \), the intensity decay time of 19 fs corresponds to a propagation length of about 5.4 \( \mu \)m, being much smaller than the size of the spot of excitation.

Theoretically, we expect a Fourier relation between the time-resolved visibility of Fig. 4 and the transmission spectrum of the hole array. The described exponential decay in time corresponds to a Lorentzian-shaped transmission spectrum with a FWHM of \( 1/\pi\tau_c \). The calculated value of 18.5 nm is indeed close to the FWHM of the

![Fig. 3. Time-resolved polarization decoherence, measured as the polarization fringe visibility \( V \) versus time delay \( \tau \), for a hole array positioned in the focus of a telescope of variable numerical aperture NA. The solid curve without markers shows the measurement without hole array as a reference. The horizontal line depicts the zero level.](image)

![Fig. 4. (Color online) Averaged absolute values of \( V(−\tau) \) and \( V(+\tau) \) in Fig. 3, plotted on a vertical logarithmic scale as a function of \( |\tau| \). The thicker curve without markers represents the measurement without hole array. The straight solid line is a fit of the exponentially decaying part of the NA=0.017 curve, from which a decay time \( \tau_c = 38\pm 1 \) fs is obtained.](image)
18 nm obtained from the transmission spectrum in Fig. 2. Because of the asymmetric Fano profile of the resonance (see Fig. 2), a more realistic model is obtained by inclusion of a $\delta(t)$ response in time (uniform background in frequency) which results in a slightly (approximately 10–20%) wider spectrum for the same decay rate.

The faster decay of the time-resolved visibility at larger numerical apertures, as shown in Figs. 3 and 4, is a result of transit time effects; surface plasmons move out of the excitation area more rapidly for large NA. Alternatively, we can interpret it in terms of a Fourier relation; the transmission spectrum becomes broader under strong focusing conditions, owing to angle-dependent spectral shifts.12

As our final and most crucial experiment, we studied the recovery of the polarization coherence by compensation of the imposed time delay by an additional delay behind the array. More specifically, we have measured the time-resolved visibility with a fixed time delay of $\tau_{\text{fix}} = 145$ fs in front of the telescope (NA=0.053) and the array (this $\tau_{\text{fix}}$ is large enough to completely remove the polarization entanglement) and a variable “reverse” time delay $-\tau_{\text{fix}} + \tau$ behind the hole array. Fig. 5 shows the measured visibility as a function of $\tau$ (solid dots) as well as the measurement without reverse time delay (open circles for $\tau_{\text{fix}}=0$ copied from Fig. 3). We note that both curves in Fig. 5 have practically the same functional shape, as the focusing conditions are equal. For the peak visibility, however, we obtain a value of $V=0.75\pm0.01$ for the reverse time-delay measurement, whereas a value of $V=0.83\pm0.01$ was found in the original measurement. In other words, the polarization decoherence induced by $\tau_{\text{fix}}$ (to $V=0$) cannot be totally compensated for by a reverse time delay $-\tau_{\text{fix}}$.

The permanent loss of polarization coherence shows that both decoherence channels are not independent, but coupled in our black box. We note that, in absence of the hole array, a peak visibility of 0.96 was measured (being a measure of the entanglement quality of our SPDC source18) thereby excluding the combination of delay–reverse delay in itself as a potential source of coherence loss.

Theoretically, the peak visibility for the reverse time-delay measurement (for large $\tau_{\text{fix}}$) can be interpreted as the average of the visibilities measured without any time delay in the 45° and the $\sigma^+$ basis. This is indeed the case, as we measured $V_{45^\circ}=0.83\pm0.01$ and $V_{\sigma^+}=0.68\pm0.01$ (see also Ref. 13), which average to $V=0.75\pm0.01$. The reason for the mentioned averaging is most easily understood in the frequency domain; within the bandwidth of our SPDC light, the polarization incident on the telescope is different for every frequency $\omega$ (owing to a varying phase delay $\omega\tau_{\text{fix}}$), i.e., it changes from $+45^\circ$, via $\sigma^+$, $-45^\circ$ and $\sigma^-$, to $+45^\circ$ again.

To confirm that the propagation of surface plasmons plays a key role in the coupling between the decoherence channels, we repeated the delay–reverse delay measurement for $\tau=0$ at low NA=0.017. We then measured a visibility of 0.90, which is much closer to its original peak value of 0.93 (see Fig. 3) than for the NA=0.053 data. This stronger recovery of polarization coherence is attributed to the slower propagation of the surface plasmons out of the larger excitation spot (see Fig. 4).

Finally, we note that our metal hole array is not as polarization isotropic as we would like, as production errors lead to some birefringence and dichroism of the array.13 To study the potential effects of these anisotropies on the (recovery of) polarization coherence, we fixed the input polarization of the upper beam in Fig. 1 by orienting the polarizer of the lower beam horizontally and used extra polarization optics to measure the Mueller matrix of the array as in Ref. 13. The measured Mueller matrix shows a comparable structure as in Ref. 13, i.e., dominant (at least 0.90) diagonal elements and finite but small ($\approx0.10$) off-diagonal elements, which quantify the slight array imperfections. By choosing convenient experimental conditions in the decoherence experiments of Figs. 3–5, we could remove most of the effects created by the off-diagonal elements. If the fringe visibilities in Figs. 3–5 would, for instance, be measured just by rotating the polarizer behind the array, we would face variations of up to $\approx 20\%$ in the single-count rate. By keeping the polarization fixed and instead varying the birefringent delay, we did not have this problem. Furthermore, as the off-diagonal elements hardly depend on the used NA,13 the drop in the measured peak visibilities $V[=M_{22}(1-M_{22})+M_{02}]$ in Fig. 3 must indeed correspond to a decrease of the diagonal element $M_{22}$ and thus to polarization decoherence and not to a mere change in the state of polarization. The above arguments show that the slight polarization-anisotropic nature of our hole array hardly affects the measured visibilities and polarization decoherence.

4. CONCLUDING DISCUSSIONS

In conclusion, we have performed time-resolved measurements of the polarization decoherence in a metal hole array under different focusing conditions. Apart from the decoherence induced by focused illumination of the hole array, we have shown that a temporal separation of the
incident orthogonal polarization components creates an additional decoherence that cannot be totally compensated for by a retiming of the polarization components after propagation through the array. This result demonstrates that the time- and space-related decoherence channels (operating on frequencies and angles, respectively) are coupled via propagating surface plasmons in a metal hole array.

An important result is that the Mueller-matrix black-box method, although convenient, should be treated with care in optical decoherence; as we have observed, it can even produce incorrect results in the analysis of a series of consecutive decoherence processes. For a complete description of the polarization evolution, beyond the simple truncated form provided by the Mueller algebra, two options are available. One option is to retain the full temporal and spatial information of the polarization. The observed coupling between the time- and space-related decoherence channels can then be mathematically explained by the noncommuting behavior of the angle-dependent transmission matrix \( t(\theta, \lambda) \) of the hole array \( \theta = (\theta_x, \theta_y) \) and the time-dependent Jones matrix \( t(\tau) \), associated with the birefringent time delay. As \( t(\theta, \lambda) \) is a nondiagonal matrix whereas \( t(\tau) \) is diagonal in the \( (H, V) \) basis (axis orientation of BBO and waveplates), it is the matrix character and not the \( \lambda \) dependence of \( t(\theta, \lambda) \) that frustrates the commutation. Another option is to divide the spatial-angular information over \( N \) discrete transverse modes. However, in this multimode description the classical evolution of our black box already requires a \( 2N \times 2N \) matrix for monochromatic incident light only. If we also include the frequency, i.e., temporal information, an even larger matrix is needed that may lead to a less transparent description.

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REFERENCES