

Deterministic and efficient source of frequency-polarization hyper-encoded photonic qubits

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The frequency or color of photons is an attractive degree of freedom to encode and distribute quantum information over long distances. However, the generation of frequency-encoded photonic qubits has so far relied on probabilistic nonlinear single-photon sources and inefficient gates. Here, we demonstrate the deterministic generation of photonic qubits hyper-encoded in frequency and polarization based on a semiconductor quantum dot in a cavity. We exploit the double dipole structure of a neutral exciton and demonstrate the generation of any quantum superposition in amplitude and phase, controlled by the polarization of the pump laser pulse. The source generates frequency-polarization single-photon qubits at a rate of 4 MHz corresponding to a generation probability at the first lens of $28 \pm 2\%$, with a photon number purity $>98\%$. The photons show an indistinguishability $>91\%$ for each dipole and 88% for a balanced quantum superposition of both. The density matrix of the hyper-encoded photonic state is measured by time-resolved polarization tomography, evidencing a fidelity to the target state of $94 \pm 8\%$ and concurrence of $77 \pm 2\%$, here limited by frequency overlap in our device. Our approach brings the advantages of quantum dot sources to the field of quantum information processing based on frequency encoding.

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Photons are versatile carriers of quantum information for applications both in quantum communications [1] or quantum computing [2]. A key advantage of photons is that they have many degrees of freedom to flexibly encode the information tailored to specific use cases. Among them, frequency has recently attracted interest [3,4], especially for quantum communication [5] since frequency is inherently stable over fiber links [6] and is compatible with current demultiplexing technology to build complex multiuser quantum networks [7]. Because of those clear advantages, frequency qubits have been used to generate secure cryptographic keys, both using weak attenuated coherent pulses [5] and entangled pairs [8,9], and to connect distant quantum memories via Bell state measurements [10,11] or Raman transitions [12,13].

Significant progress has been made toward the generation and control of photonic qubits in the frequency domain. Sources of photonic frequency qubits have relied on probabilistic sources

based on parametric downconversion and four-wave mixing that are intrinsically limited in efficiency by the presence of multi-photon components [14]. Deterministic single-qubit gates [15] as well as two-qubit gates [16] have been demonstrated using nonlinear crystals, electro-optic modulators, and pulse shapers [17,18] or electro-optic effects [15], as well as Hong–Ou–Mandel interference [17,18]. These results lay the foundations for a complete architecture for quantum information processing in the frequency domain. However, the main challenges to real-world applications remain the losses introduced for performing those gates and the probabilistic nature of the sources.

In this work, we take a meaningful step to address one of these core challenges and demonstrate the deterministic and efficient generation of polarization and frequency hyper-encoded qubits. Our approach exploits semiconductor quantum dots (QDs) [19] that are an excellent platform for the on-demand generation of

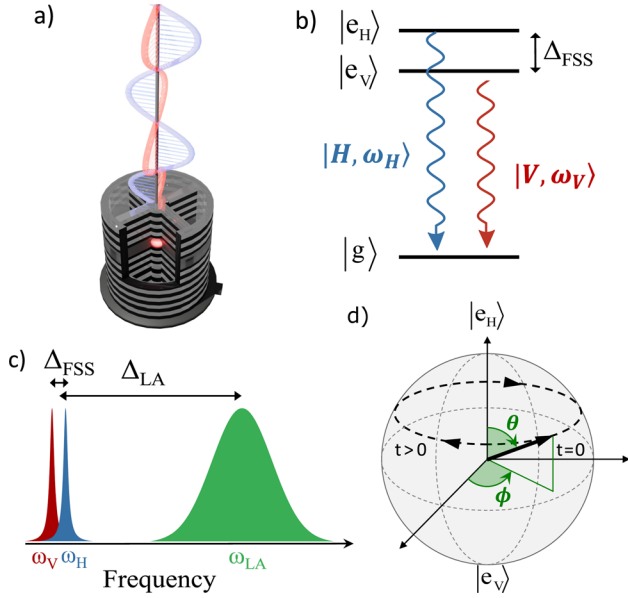


Fig. 1. Principle of the experiment. (a) Artistic visual of the device, a semiconductor quantum dot (red) embedded in a micropillar cavity, generating a photonic qubit hyper-encoded in the frequency and polarization degrees of freedom. (b) Energy levels of the neutral QD, comprising one ground state and two excited states separated in energy by the fine structure splitting Δ_{FSS} and emitting orthogonal, linearly polarized photons. (c) Sketch of the excitation technique. A near-resonant laser pulse (green) with detuning $\Delta_{LA} \gg \Delta_{FSS}$ can excite the two dipole transitions via a phonon-assisted process, either individually or in a superposition. (d) Bloch sphere representation of the QD in the excited state. The parameters θ and ϕ (vector at $t = 0$) are controlled by the pump laser polarization. The energy difference between the two states Δ_{FSS} induces a coherent evolution of the photonic state for $t > 0$.

pure and indistinguishable single photons [20–22]. We exploit the energy levels of the neutral QD embedded into a micropillar cavity, which we address via an efficient, off-resonant excitation scheme assisted by the relaxation of longitudinal acoustical (LA) phonons [23]. In addition to the superposition in the frequency domain, the two single-photon modes are also orthogonal in polarization, a feature leading to non-classical correlations between these two degrees of freedom (intra-photon entanglement [24]). We perform time-resolved polarization tomography that allow us to measure the density matrix of the intra-entangled state and characterize the photonic qubit source in terms of efficiency, single-photon purity, and indistinguishability.

The device under study is illustrated in Fig. 1(a). It consists of an InGaAs QD deterministically embedded in a micropillar structure surrounded by distributed Bragg reflectors [25] and electrically contacted [26]. The device is placed in a closed-cycle cryostat and cooled down to 8 K. Single photons are generated and collected using a confocal microscopy setup [23]. Figure 1(b) shows the energy levels of the neutral QD, comprising a three-level structure with one ground state $|g\rangle$ and two excited states $|e_H\rangle$ and $|e_V\rangle$ separated in energy by the fine-structure splitting (Δ_{FSS}). This splitting of typically few to tens of μeV in InGaAs QDs arises from the natural asymmetry in self-assembled quantum dots [27] due to the strain-induced growth process. Another consequence of this anisotropy is that the two optical transitions occur with linear, orthogonal polarizations

denoted $|H\rangle$ and $|V\rangle$. In this work, we make use of these two dipole transitions as the eigenmodes of a photonic qubit.

We generate single photons from the QD using the longitudinal acoustic (LA)-phonon-assisted excitation scheme [23,28] depicted in Fig. 1(c). In this scheme, a 15-ps-long laser pulse, blue-detuned from the QD central wavelength by $\Delta_{LA} = 0.65$ nm, can excite both dipoles with similar efficiencies since $\Delta_{LA} \gg \Delta_{FSS}$. The strong excitation pulse first dresses the ground and excited states, enabling the fast (few ps) relaxation of an LA phonon between the dressed states. Adiabatic undressing of the energy levels at the end of the laser pulse then leaves the QD in its excited state with high probability, which then recombines through spontaneous emission of a photon. Despite being an incoherent process, this scheme has been demonstrated to yield an efficient population inversion along with high levels of single-photon purity and indistinguishability [23]. It also faithfully preserves the polarization of the selection rules and its broadband nature allows exciting both dipoles [29], a crucial feature for this work. The detuning Δ_{LA} between the pump laser pulse and the QD then allows to spectrally filter the pump laser pulse, allowing to excite and collect photons with arbitrary polarization. In particular, the polarization of the pump laser can be set either to address selectively one of the two transitions or to generate a superposition of the two dipoles.

The general case is depicted in Fig. 1(d). The excitation laser pulse has polarization $|\theta, \phi\rangle = \cos(\frac{\theta}{2})|H\rangle + \sin(\frac{\theta}{2})e^{i\phi}|V\rangle$, where $\frac{\theta}{2}$ is the angle between the laser polarization and the H dipole of the QD, and ϕ accounts for the ellipticity of the pulse ($\phi = 0$ corresponding to linear polarization). This pulse prepares the state $|\psi_{QD}\rangle$ of the QD in a superposition of the two excited states $|e_H\rangle$ and $|e_V\rangle$ corresponding respectively to the horizontally and vertically polarized dipoles at $t = 0$, which evolves until decay according to the quantum trajectory

$$|\psi_{QD}(t)\rangle = \cos\left(\frac{\theta}{2}\right)e^{-i\omega_H t}e^{-\frac{t}{\tau}}|e_H\rangle + \sin\left(\frac{\theta}{2}\right)e^{-i(\omega_V t - \phi)}e^{-\frac{t}{\tau}}|e_V\rangle, \quad (1)$$

where ω_H (ω_V) is the central frequency of the horizontally (vertically) polarized dipole and τ is the lifetime of the excited state. This temporal evolution of the excited state of the QD is then imprinted on the state of the emitted photon $|\psi\rangle$ as a consequence of the proportionality relation between the QD dipole operator and the field operator of the electromagnetic mode. In the absence of dephasing, the photonic state can thus be written as

$$|\psi\rangle = \cos\left(\frac{\theta}{2}\right)|H, \omega_H\rangle + \sin\left(\frac{\theta}{2}\right)e^{i\phi}|V, \omega_V\rangle, \quad (2)$$

where the pulse mode $|\omega_j\rangle$ is

$$|\omega_j\rangle = \frac{1}{\sqrt{\tau}} \int_0^\infty e^{-i\omega_j t} e^{-\frac{t}{\tau}} \hat{a}^\dagger(t) |0\rangle dt \quad (3)$$

in the time basis, for polarization $j \in \{H, V\}$.

We remark that, provided $\Delta\omega = \omega_H - \omega_V = \Delta_{FSS}/\hbar$ is nonzero, the state in Eq. (2) is non-separable: the frequency and polarization degrees of freedom of a single emitted photon are entangled, a feature referred to as *intra-particle* entanglement [30] or *single-particle* entanglement [24]. Such a feature can be exploited in quantum information protocols for instance in robust quantum communication [31], efficient quantum logic

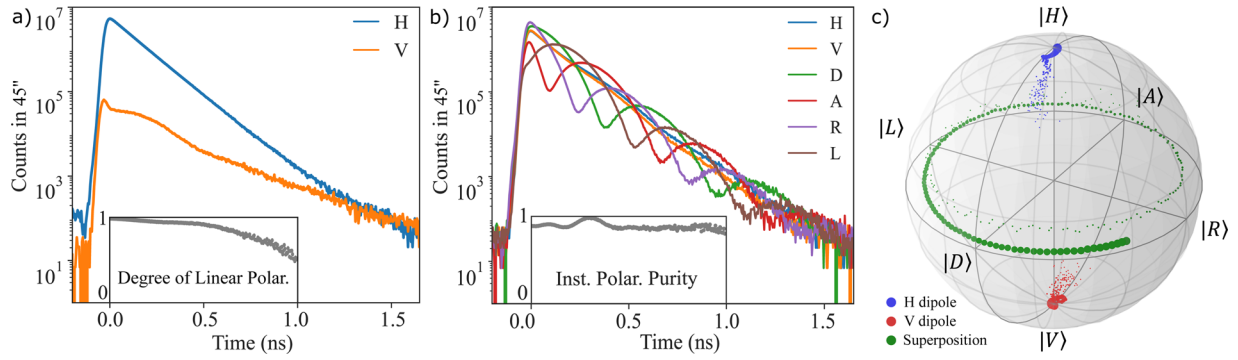


Fig. 2. Time-resolved polarization tomography of the generated quantum states. Time traces (a) for a pure state (H dipole) measured in the H/V basis and (b) for a frequency-polarization hyper-encoded photonic qubit, measured in all three bases (full tomography). The insets show respectively the degree of linear polarization and instantaneous purity over time between 0 and 1 ns (linear scale). (c) Trajectory of the photonic qubits in the Bloch sphere. Point sizes are weighted by their intensity.

[32–35] or random number generation [36]. Alternatively, it can be removed in a deterministic manner [37]. In the following, we characterize the polarization and frequency hyper-encoding by performing polarization-resolved measurements in the time domain. For this, we use superconducting nanowire single-photon detectors (SNSPDs) with temporal resolution below 30 ps $\ll 2\pi/\Delta\omega$, allowing us to resolve the temporal evolution described in Eq. (1).

We first generate single photons in a pure state for each dipole, $|H, \omega_H\rangle$ or $|V, \omega_V\rangle$. The emitted photons are sent through a polarization tomography stage and detected by SNSPDs. We set the polarization of the exciting laser parallel to the H dipole and record the time traces of the emission in polarization parallel (H) and perpendicular (V) to the dipole. The obtained time traces are shown in Fig. 2(a). We observe a mono-exponential decay, giving a radiative lifetime of $\tau = 133$ ps, highly polarized along the dipole polarization. The inset displays the degree of linear polarization (D_{LP}) of the emission over time, between 0 and 1 ns, (or equivalently, the Stokes parameter in the HV basis, S_{HV}) defined as

$$D_{LP}(t) = S_{HV}(t) = \frac{I_H(t) - I_V(t)}{I_H(t) + I_V(t)}. \quad (4)$$

Integrating over the full wavepacket, we measure $D_{LP} = 97.1\%$ (weighted by the instantaneous fluorescence intensity), while the instantaneous D_{LP} slightly decreases over time indicating the presence of dissipative processes in the excited state. We perform the same measurement with the pump laser aligned along the V dipole (data in Supplement 1), measuring a time average $D_{LP} = 96.8\%$ for this dipole.

We now set the polarization of the pump laser to excite the two dipoles with equal probability, and record the time traces of the emission, this time along all six polarization axes (full polarization tomography). The results are displayed in Fig. 2(b). We observe monoexponential decays in the H and V bases with equal intensities, whereas coherent oscillations appear in the D/A/R/L polarizations. Such time traces can be understood by projecting the photonic wavepacket of Eq. (2) along the corresponding polarization measurement bases. In particular, performing a measurement along the D basis gives the measured intensity:

$$I_D(t) = \frac{1}{2} \langle \psi | (\hat{a}_H(t) + \hat{a}_V(t))^\dagger (\hat{a}_H(t) + \hat{a}_V(t)) | \psi \rangle \\ = \frac{1}{2\tau} e^{-\frac{t}{\tau}} (1 + \cos(\Delta\omega t - \phi) \sin(2\theta)), \quad (5)$$

with $a_j^\dagger(t) = a^\dagger(t)|j\rangle$ for polarization $j \in \{H, V\}$. Similar expressions can be derived for the A/R/L bases, whereas the time trace following a projective measurement in the eigenstates bases H and V result only in a monoexponential decay. From Eq. (5), we deduce that the period of the oscillations provides a direct measurement of the fine structure splitting and obtain via a fit of the time trace $\Delta_{FSS} = \hbar\Delta\omega = 7.35$ μ eV. This value is consistent with values obtained for annealed QDs studied here where strain and anisotropy are reduced.

Using the time traces from Fig. 2, we reconstruct the Stokes vector of the state over time, defined as $S = (S_{HV}, S_{DA}, S_{RL})$ with S_{DA} and S_{RL} defined similarly as in Eq. (4). We plot the state evolution over time in the Bloch sphere in Fig. 2(c) (green points), where we observe a coherent evolution around the H/V axis, as described in Fig. 1(d). For comparison, in blue data from Fig. 2(a) and red data in Supplement 1 are the eigenstates generated when exciting along the dipoles H and V.

The inset in Fig. 2(b) shows the instantaneous polarization purity over time, defined as the norm of the Stokes vector. A high purity is observed with hardly any decrease over the lifetime, with an integrated purity of 90.0% over the total wavepacket (weighted by the instantaneous fluorescence intensity). The rather constant polarization purity over time seems in contradiction with the decrease in the DLP for the pure state (Fig. 2(a) inset). We attribute this observation to the residual population of the biexciton state in the case of linearly polarized excitation (Fig. 2(a)). Since populating the biexciton state is only efficient with linearly polarized excitation (due to spin-polarized selection rules), its influence on the generated state in Fig. 2(b) is likely to be minimal if the excitation polarization is close to circular.

To demonstrate that we can generate such a state with arbitrary initial amplitude and phase, we now vary the pump laser polarization (angles θ_{laser} and ϕ_{laser}) and perform similar time-resolved polarization tomography measurements for 10 different sets of excitation polarization angles in total. From the time traces, we retrieve the values of the angles θ and ϕ for the generated state at $t = 0$, which we display in the Bloch sphere in Fig. 3 along with the numerical values. The points span across the entire Bloch sphere, including equal and unequal superpositions with arbitrary initial phase. The exact correspondence between the excitation parameters and the amplitude and phase of the generated state is affected by the birefringence of the optical components in the laser path; however, it is possible

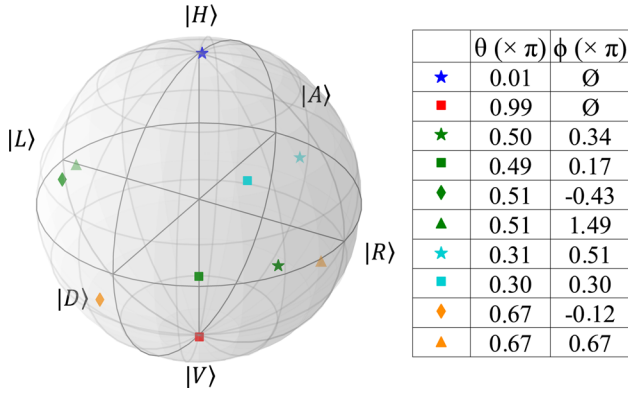


Fig. 3. Generation of any arbitrary superposition between the two dipoles. The laser polarization is varied to generate 10 states with different initial amplitudes and phases. The fits of the time-resolved polarization tomography for each state allow to extract the values of θ and ϕ (as defined in Fig. 1(d)), which are displayed in the legend. The shaded symbols (green triangle, cyan star, orange triangle) are located at the back of the sphere.

to characterize and compensate for this unitary transformation introduced. We note that in the general case, the photonic state is described by a vector in a composite Hilbert space with two dimensions of frequency and two of polarization, whose basis is constituted by the four possible single-photon states $|H, \omega_H\rangle$, $|V, \omega_V\rangle$, $|H, \omega_V\rangle$, and $|V, \omega_H\rangle$. While our scheme allows to generate arbitrary superpositions of the first two basis states, the other two are accessible by simple unitary operations on the polarization of the generated photons, i.e., with a set of waveplates.

We now turn to the analysis of the single-particle entanglement arising from the non-separability between polarization and frequency in Eq. (2). Using the polarization-resolved time traces for a given generated state (Fig. 2(b)), we can retrieve the coefficients of its density matrix in the composite Hilbert space. We derive the expressions of the expected intensities over time after projecting the wavepacket described in Eq. (2) along the six measurement axes (detailed in Supplement 1), and obtain a set of equations which we fit to the time traces, using a maximum likelihood method [38]. This allows us to estimate the coefficients of the density matrix. We show in Fig. 4 the result obtained in the case of the green triangle in Fig. 3 after a change of basis to remove the imaginary part in the result (detailed in Supplement 1). The obtained density matrix is close to the expected one for the target state $|\phi^+\rangle = \frac{1}{\sqrt{2}}(|H, \omega_H\rangle + |V, \omega_V\rangle)$, with estimated fidelity $F = 94 \pm 8\%$. Our approach could also generate the orthogonal Bell state $|\phi^-\rangle = \frac{1}{\sqrt{2}}(|H, \omega_H\rangle - |V, \omega_V\rangle)$ by exciting with the orthogonal polarization, while the other two Bell states $|\psi^\pm\rangle = \frac{1}{\sqrt{2}}(|H, \omega_V\rangle \pm |V, \omega_H\rangle)$ can be obtained by combining the scheme with polarization unitary operations.

Finally, we assess the performance of the device as a single-photon source, namely in terms of efficiency, single-photon purity, and indistinguishability. In Fig. 2(a), the total detected rate for the H dipole is 4.7 MHz, for a repetition rate of the laser $R_L = 82$ MHz. Considering optical losses in our experimental setup (see Supplement 1), this corresponds to a first lens brightness value of $\mathcal{B}_{FL} = 28 \pm 2\%$. A slightly lower detected count rate of 3.4 MHz is found for the V dipole, (data in Supplement 1) corresponding to a first lens brightness of $21 \pm 2\%$. The difference in brightness between the two dipoles can be attributed to

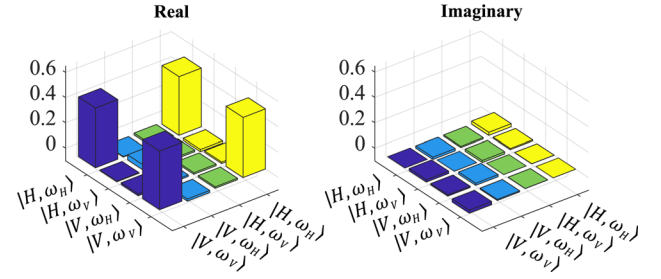


Fig. 4. Intra-entanglement between the polarization and frequency degrees of freedom of a single photon. Reconstructed density matrix of a hyper-encoded state in frequency and polarization. The fidelity to the target state is estimated at $F = 94 \pm 8\%$.

valence-band mixing in C_{2v} symmetry [27]. Note that several QD-based devices have already demonstrated brightness values above 50% [20], including a first-lens brightness of 55% in the LA phonon-assisted scheme [21], setting an estimation of the achievable rates.

The second-order intensity correlation gives $g^{(2)}(0)$ as low as $g^{(2)}(0) = 1.70 \pm 0.02\%$ and $g^{(2)}(0) = 1.47 \pm 0.01\%$ respectively for each dipole, and $g^{(2)}(0) = 1.3 \pm 0.1\%$ for a balanced superposition of the two dipoles (see Supplement 1), demonstrating the single-photon nature of the source. The indistinguishability is measured via the Hong–Ou–Mandel interference of the full wavepacket, without filtering the photons with a polarizer. We obtain respectively $V_{HOM} = 88.9 \pm 0.3\%$ and $88.8 \pm 0.3\%$ for the pure dipoles H and V, corresponding to indistinguishabilities of $M = 92.2 \pm 0.1\%$ and $91.6 \pm 0.1\%$ [39] as shown in Fig. 5, which further increase to $M_H = 94.5 \pm 0.1\%$ and $M_V = 93.8 \pm 0.1\%$ if the photons are filtered with a polarizer. For a balanced quantum superposition of the two dipoles, we measure the full wavepacket overlap, comprising the two orthogonally polarized modes. We obtain $V_{HOM} = 85.9 \pm 0.1\%$, corresponding to $M = 88.3 \pm 0.1\%$. This value is only slightly lower than the one obtained for the eigenstates, in line with the integrated polarization purity (inset of Fig. 2) and further evidences the coherence of the state generated.

The dashed lines and the black line in Fig. 5 display the expected interference visibilities predicted by our model, taking into account a dissipative mixing of the $|e_H\rangle$ and $|e_V\rangle$ excited states, a slight imbalance between the two measured dipole magnitudes, and an initial state preparation impurity (detailed in Supplement 1). The dashed lines show that the polarization-filtered visibilities drop substantially as the excitation polarization becomes orthogonal to the filtered polarization. This is caused by the filter progressively allowing proportionally more photons through that arise following a polarization flip in the QD excited state, and hence the light becomes more incoherent. The fit of the expected visibility (black line) to the unfiltered visibility measurements yields a time scale for this dissipative mixing of 12 ± 1 ns. The difference between the unfiltered values at $\theta = 0$ and $\theta = \pi/2$ is explained by a V-polarized dipole that is approximately 12% weaker, which qualitatively agrees with the observed difference in the count rates. We also find that the reduced mean wavepacket overlap given by the superposition state implies an initial state preparation purity of $94.9 \pm 0.2\%$, which corresponds to the observed non-unity instantaneous polarization purity at t_0 (see inset of Fig. 2(b)).

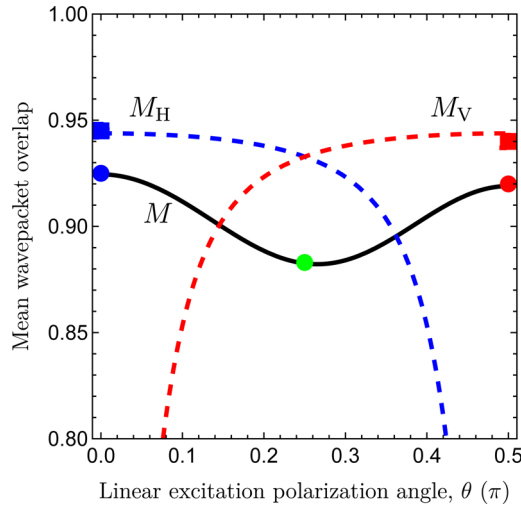


Fig. 5. Mean wavepacket overlap M obtained from the HOM interference visibility, measured for the H (blue, $g^{(2)}(0) = 1.47\%$, $M = 91.6 \pm 0.1\%$) and V (red, $g^{(2)}(0) = 1.47\%$, $M = 92.2 \pm 0.1\%$) dipoles, as well as for an equal superposition of the two dipoles (green, $g^{(2)}(0) = 1.7\%$, $M = 88.3 \pm 0.1\%$). The circles are measured without polarization projection, while the squares are the values M_H and M_V taken after the photons are filtered with a polarizer. The black line shows the expected value from our model (see text), and the dashed lines correspond to the simulated values of M_H and M_V .

An important parameter in our approach is the QD fine structure splitting, which governs the overlap between states $|\omega_H\rangle$ and $|\omega_V\rangle$ by $|\langle\omega_H|\omega_V\rangle|^2 = 1/(1 + \Delta\omega^2\tau^2)$ following Eq. (3). The relatively small splitting in our devices based on annealed QDs results in a non-negligible overlap between the two dipoles: considering the radiative lifetime of 133 ps and the energy splitting of $\Delta_{FSS} = 7.35 \mu\text{eV}$, we calculate an overlap of 31.2%. We have experimentally verified this value via the HOM interference between the two dipoles [40] (see Supplement 1), for which we obtained a value of $M_\omega = 31.8 \pm 0.1\%$ in good agreement. The overlap also dictates the amount of intra-particle entanglement. By applying a Gram–Schmidt orthogonalization to Eq. (2) (see Supplement 1), we find that the polarization-frequency entanglement concurrence is bounded from above by $C \leq \sqrt{1 - |\langle\omega_H|\omega_V\rangle|^2} \simeq 83\%$. Using the measured overlap from HOM interference $|\langle\omega_H|\omega_V\rangle|^2 \simeq M_\omega$, and by applying the same orthogonalization process to the reconstructed density matrix, provides an entanglement concurrence of $C = 77\%$.

An FSS of 200 μeV , that is reasonably achievable for InAs QDs would result in an overlap of 0.06%, allowing to properly resolve the two frequency modes and providing an entanglement concurrence as high as 99.97%. Moreover, we note that the FSS of a quantum dot can be made tunable by engineering the device. This has been an extensive topic of research in the context of entangled photon pair generation with neutral QDs [41,42], and was demonstrated using strain [43], magnetic field [44], or in-plane electric fields [45].

In conclusion, we have reported on an efficient approach to generate hyper-encoded single photons in a superposition of their frequency and polarization degrees of freedom in a deterministic manner. Our device, a semiconductor quantum dot in a microcavity, brings near-deterministic single-photon emission at high rates to the field of frequency encoding. The

emitted single photons show high purity and indistinguishability, enabling them to perform quantum interferences for quantum information processing applications.

Going further, the present scheme could be transferred to other QD materials, such as droplet GaAs QDs for interfacing with quantum memories [46] or InP QDs for telecommunication wavelength operation [47]. We emphasize that the inhomogeneous distribution in frequency between various QDs could be leveraged to reach a large number of frequency modes, by combining the present scheme with several QD devices and frequency gates [3]. Finally, the scheme could be extended to generate spin-entangled frequency qubits [48] using the four non-degenerate optical transitions of a charged quantum dot in a high magnetic field, or even to larger dimensions using magnetic quantum dots [49].

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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