

# Scanning cavity microscopy of a single-crystal diamond membrane

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(Dated: 12 October 2022)

Spin-bearing color centers in the solid state are promising candidates for the realization of quantum networks and distributed quantum computing. A remaining key challenge is their efficient and reliable interfacing to photons. Incorporating minimally processed membranes into open-access microcavities represents a promising route for Purcell-enhanced spin-photon interfaces: it enables significant emission enhancement and efficient photon collection, minimizes deteriorating influence on the quantum emitter, and allows for full spatial and spectral tunability, key for controllably addressing suitable emitters with desired optical and spin properties. Here, we study the properties of a high-finesse fiber Fabry-Pérot microcavity with integrated single-crystal diamond membranes by scanning cavity microscopy. We observe spatially resolved the effects of the diamond-air interface on the cavity mode structure: a strong correlation of the cavity finesse and mode structure with the diamond thickness and surface topography, significant transverse-mode mixing under diamond-like conditions, and mode-character-dependent polarization-mode splitting. Our results reveal the influence of the diamond surface on the achievable Purcell enhancement, which helps to clarify the route towards optimized spin-photon interfaces.

## I. INTRODUCTION

Quantum networks built from individual optically addressable spins in solids interfaced with single photons<sup>1,2</sup> promise a variety of emerging applications, ranging from secure communication over large distances to distributed quantum computing, which could become the basis of a future quantum internet<sup>3</sup>. A key building block for this is an efficient interface between the spin and photons to enable deterministic transfer of quantum states between stationary and flying qubits<sup>4-7</sup>. The most powerful approach in this respect is to couple color centers to optical microcavities and harness the Purcell effect to enhance the emission<sup>8,9</sup>. This reshapes the emission pattern into a single, well collectable mode, increases the emission fraction into the coherent zero-phonon line (ZPL), and broadens the transition due to the shortening of the excited state lifetime such that spectral fluctuations can be masked to improve photon indistinguishability.

Nitrogen-vacancy (NV) centers in diamond are a prime candidate material system in this respect that stands out due to its exceptional spin coherence properties, the availability of a nuclear spin quantum register<sup>10</sup>, and lifetime-limited optical transitions that permit the generation of spin-photon entanglement<sup>11</sup>. Several cavity architectures have been investigated to demonstrate the basic principle of Purcell enhancement of NV center emission<sup>12-18</sup>. Due to the high sensitivity of NV centers to fluctuating electric fields, a promising approach to achieve narrow optical transitions

inside a cavity is based on open-access Fabry-Pérot microcavities with incorporated minimally processed single-crystal membranes<sup>19-24</sup>, see figure 1a). This approach has led to first successful experiments, both with NV<sup>21,24</sup>, SiV<sup>25,26</sup> and GeV centers<sup>27</sup> in diamond, as well as with rare earth ions in oxide crystals<sup>28</sup> recently, and Purcell enhancement could be demonstrated. However, the desired net improvement of the collection of lifetime-limited NV center ZPL photons has not been achieved to date. This emphasizes the further need to understand limitations and improve the experimental realization of such systems.

One critical aspect lies in the details of the diamond membrane and its influence on the cavity modes. The presence of the diamond-air interface leads to a hybridized mode structure<sup>6,19,23,29</sup>. Depending on the diamond thickness, the cavity standing wave light field can either fulfill the boundary condition for the air gap part where a field node should form at the interface, called an air-like mode, or match for the mode in the diamond part where a field maximum at the interface is present, a so-called diamond-like mode, as shown in figure 1 (d) and (e). Since both conditions cannot be met simultaneously, hybridized modes form, whose dispersion, finesse and loss strongly depend on the mode character<sup>19,29</sup>. For such a hybridized diamond-air cavity, the mode dispersion, i.e. the frequency dependent shift of the cavity resonances, deviates from the linear behavior known from a bare Fabry-Pérot cavity and shows a varying slope for different diamond thickness at a fixed wavelength. As shown in a simulation of cavity

resonances in figure 1 (f), the dispersion exhibits the smallest slope for a configuration with predominant diamond-like character, and the steepest slope for a predominant air-like configuration as in figure 1 (g). To maximize the coupling to color centers in the membrane, diamond-like modes are beneficial because they show a larger electric field inside the diamond<sup>29</sup> as it can be seen by comparing figures 1 (d) and (e). However, for this configuration, the scattering loss at the diamond surface becomes maximal, requiring a trade-off depending on the surface roughness level. Furthermore, a recent study observed additional loss associated with diamond-like modes which could not be conclusively explained by surface scattering<sup>30</sup>. In general, a comprehensive investigation of the cavity performance and its relation to the diamond surface topography would be desirable, while experiments to date report only punctual measurements of cavity performance on diamond membranes, mostly under air-like conditions.

In this work, we perform a systematic study of the cavity performance in the presence of diamond membranes. We use scanning cavity microscopy<sup>31–33</sup> at a high finesse of up to 20000 to probe extended areas of a membrane. We observe a spatial variation of the cavity mode character which follows the diamond thickness profile, a corresponding cavity transmission and finesse variation, and the local influence of defects. In addition, we identify spatially localized mode mixing<sup>34,35</sup>, mostly present in diamond-like regions, which renders a large part of these areas useless for cavity experiments. We model the observed losses based on surface scattering, absorption, and mirror transmission, and find that we need to assume a larger surface roughness than measured. In addition, we find that the model reproduces the data better when assuming a significant bulk absorption in the diamond. Furthermore, we observe an increase of the cavity loss for air-like modes with increasing diamond thickness which is larger than explainable by absorption loss. Finally, we study the polarization modes of the cavity and observe a systematic dependence of the polarization mode splitting on the mode character, as well as a background birefringence. The latter shows a spatial variation of its strength and orientation, which we interpret as a signature of the local strain distribution in the material. The results show that while a dominant fraction of the diamond membrane is free from intrinsic defects or excessive roughness-induced scattering loss, mode mixing and polarization mode splitting can lead to additional limitations. The increased loss reported in<sup>30</sup> and<sup>19</sup> appear to be consistent with our findings. We suggest that the mismatch between the mode’s curved phase front and the planar diamond-air interface can be the origin. We remark that the observed loss and birefringence effects are observed and predicted to become smaller for thinner diamond membranes. This suggests that one can target a compromise between improved NV coherence for thicker and improved cavity performance for thinner diamond membranes to achieve an optimized spin-photon interface.

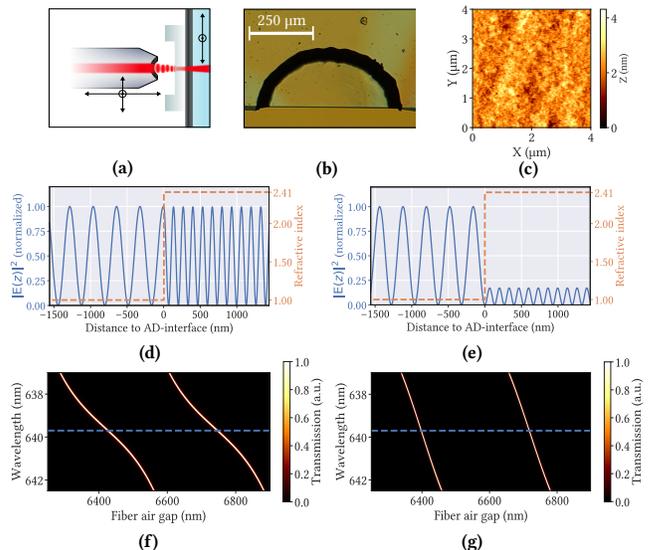


FIG. 1. (a) Schematic drawing of the cavity setup: A planar mirror (blue) carrying a diamond membrane can be moved laterally to select a region of interest. A cavity is formed together with a fiber mirror that can be nano-positioned along three axes to record raster-scanning images. (b) Microscope image of a bonded membrane. A thinned-down part for subsequent characterization lies within the region framed by the semicircle shadow. (c) AFM measurement of the membrane showing an rms-roughness of  $\sigma_{\text{rms}} = 0.4 \text{ nm}$ . (d) and (e) Simulation of the electric field inside the cavity for a diamond-like (d) and an air-like (e) mode. The refractive index profile (orange dashed line) highlights the air-diamond (AD) interface. (f) and (g) Simulated cavity resonance frequency as a function of the mirror separation for a diamond thickness of 6035 nm (diamond-like) and 6103 nm (air-like), respectively. The probe wavelength of subsequent measurements  $\lambda = 639.7 \text{ nm}$  is indicated by a dashed blue line.

## II. METHODS AND MATERIALS

Our experiments are performed using a fiber-based Fabry-Pérot cavity which is schematically illustrated in figure 1 (a). It consists of a macroscopic plane mirror and a concave mirror that is processed at the end facet of an optical fiber by  $\text{CO}_2$ -laser machining<sup>36</sup>. We use two different fiber tips  $F_A$  ( $F_B$ ) that show concave profiles with small ellipticity, characterized by radii of curvatures (ROCs) of 33.1 (55.2)  $\mu\text{m}$  along one half axis and 30.5 (48.7)  $\mu\text{m}$  along the orthogonal one. The fiber end facets are coated by ion beam sputtering with a distributed Bragg reflector (DBR) (Laseroptik GmbH, Garbsen, Germany), designed for a center wavelength at 637 nm. Numerical simulations using a transfer matrix model along with the measured layer thicknesses of the DBR stacks yield transmissions of 52 (57) ppm for the fiber mirrors at a wavelength of  $\lambda = 639.7 \text{ nm}$  that is used for the experiment. The plane mirror ( $M_A$ ) consists of a superpolished fused silica substrate ( $\sigma_{\text{rms}} < 0.2 \text{ nm}$ ), coated for a transmission of 57 ppm. More details on the cavity mirrors as well as the expected and measured finesse of the assembled cavities without the presence of a diamond sample can be found in the supplementary ma-

terial.

We study two CVD-grown single-crystal diamond samples of general grade (*Cornes Technologies, San Jose, CA, USA*) and electronic grade (*element 6*) quality. The main sample of this study (general grade quality) is structured using an inductively-coupled plasma reactive ion etching (ICP-RIE) procedure, resulting in a membrane with a minimal thickness of approximately  $5\ \mu\text{m}$ , as described in<sup>23</sup>. Characterization on  $4 \times 4\ \mu\text{m}^2$  sub-regions of the membrane with an atomic force microscope (AFM) - as exemplary shown in figure 1 (c) - reveals a surface roughness of  $\sigma_{\text{rms}} = 0.4 - 0.5\ \text{nm}$  after the final etching steps. For bonding of the membranes onto plane mirrors we exploit van der Waals forces between the mirror and the sample. Initial interference fringes observed under a light microscope, indicating an air gap between the membrane and the mirror, broaden during the bonding procedure, and the final result for the general grade sample depicted in figure 1 (b) shows almost no fringes (see<sup>23</sup> for details on the bonding procedure).

We use a custom-developed nanopositioning stage to control the cavity. It can be built cryo-compatible and achieves very high passive stability and fast scanning speed<sup>37</sup>. In order to select a position on the a sample, the plane mirror can be moved laterally, while the fiber can be scanned additionally along all three dimensions using piezo-electric actuators to perform the scanning cavity measurements. To probe the cavity, we couple light of a tuneable diode laser (TDL) (*Toptica DL pro 637*) at a wavelength of  $\lambda = 639.7\ \text{nm}$  into the cavity fiber and measure the transmission behind the plane mirror using an avalanche photodiode (APD) (*Thorlabs APD130A2/M*) connected to a 12-bit digital storage oscilloscope. To obtain spatially resolved maps, we raster-scan the fiber laterally on an area of about  $60 \times 60\ \mu\text{m}^2$  over the sample and modulate the cavity length at each position over multiple free spectral ranges (FSR) at a rate of 200 Hz. The maximum transmission is recorded at each position, leading to a 2D-image that shows relative changes of the cavity transmission as a function of the position on the membrane, which we refer to as a cavity transmission scan. To obtain the cavity finesse, we modulate the cavity length at a reduced rate of typically 20 Hz and probe two consecutive fundamental resonances, which we fit with Lorentzian lines. From this we calculate the finesse from the ratio of the resonance distance and the average full width at half maximum (FWHM). Due to piezo nonlinearity and hysteresis, such measurements have an uncertainty of  $\approx 10\%$  unless calibrated with care. For these measurements, we align the polarization of the input light with one of the two cavity polarization modes to probe an isolated resonance.

### III. RESULTS AND DISCUSSION

#### A. Mode-character-dependent cavity loss

We study the spatially varying effect of the diamond membrane on cavity modes by performing laterally resolved cavity transmission measurements as described above. To increase the imaged area, 16 of such scans are performed at neigh-

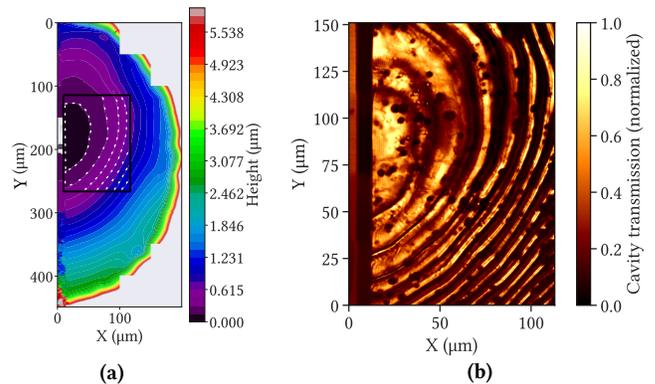


FIG. 2. (a) Surface topography of the etched diamond membrane. The black frame indicates the region where cavity transmission scans are performed, white dashed lines are used to highlight the contour lines inside this region. (b) Cavity transmission map of the framed region in (a).

boring positions on the membrane with slight overlaps at the edges and consecutively stitched together.

The final transmission scan obtained with fiber mirror  $F_A$  is shown in figure 2 along with a height map of the membrane taken with a home-built white-light interferometric microscope (WLI). Notably, the cavity transmission in figure 2 (b) exhibits distinct bright and dark fringes that match the shape of the height map in figure 2 (a), evidencing the correlation between the cavity transmission and the membrane thickness. Since the composition of the cavity mode changes with the diamond thickness at a periodicity of  $\lambda / (2n_d)$ <sup>29</sup>, with  $n_d = 2.41$  the refractive index of diamond, our measurements reveal the relative change of the cavity transmission as a function of the hybridized mode composition. To compare the thickness change of the diamond membrane obtained from the WLI measurement to the structure observed in cavity transmission, we estimate the region covered by the cavity scans (see supplementary material). This region is indicated by the black frame in figure 2 (a). For a horizontal line at the central y-position of this region, the WLI image shows a thickness change of  $\Delta_h^{\text{WLI}} \approx 770\ \text{nm}$ . Roughly seven bright fringes along the x-axis in the central y position of the cavity scan yield a thickness change of  $\Delta_h^{\text{cav}} = (7 - 1) \cdot \lambda / (2n_d) \approx 800\ \text{nm}$ , thus showing good agreement with the WLI measurement.

To understand how the composition of the hybridized cavity mode affects the cavity losses in more detail, we proceed with measuring the cavity finesse and the mode composition for points along a horizontal line, covering several bright and dark fringes in figure 3 (a). As shown in figure 3 (b), the cavity finesse spans values between 200 and 5500 and follows closely the cavity transmission measurements. In order to link each measurement position with the character of the hybridized mode, we investigate the dispersion of the cavity mode. To this end, we couple light from a white-light laser (*Fianium Whitelase SC450*) into the cavity and measure transmission spectra with a spectrometer (*Andor Shamrock*) for different cavity lengths. The mode character is derived from the slope of the mode dispersion at the wavelength  $\lambda = 639.7\ \text{nm}$

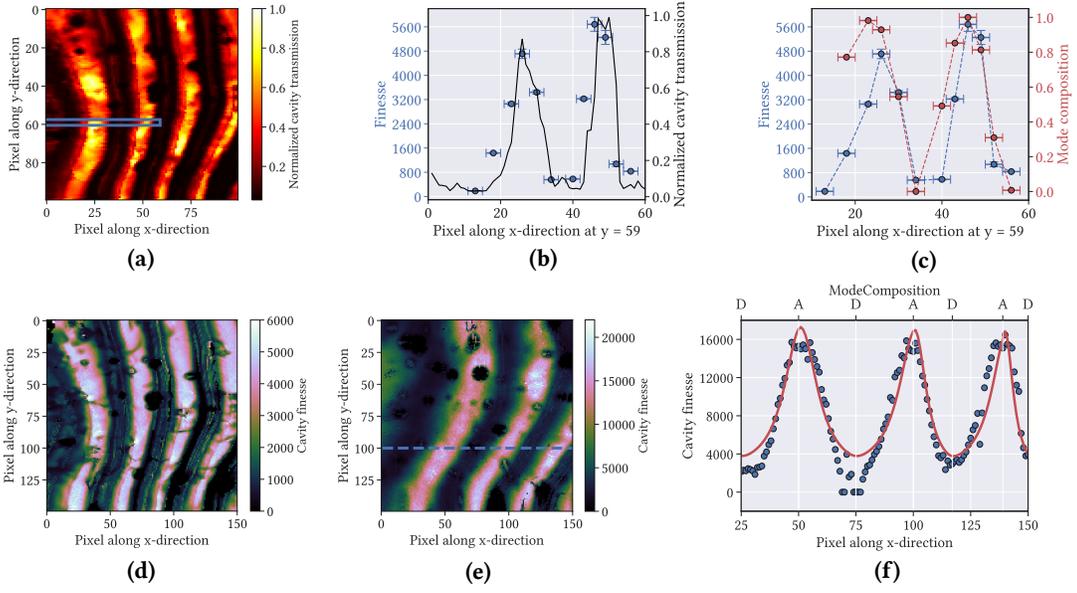


FIG. 3. (a) Cavity transmission scan on the membrane. Each pixel shows the maximum cavity transmission from several fundamental mode orders. (b) Finesse of the cavity for points along the blue line in (a). Each data point shows the average of 50 measurements. The black curve shows the transmission data from (a). (c) Cavity finesse (data from (b)) and composition of the hybridized mode obtained by measuring the mode dispersion for each point. (d) Cavity finesse scan on the same region. (e) Cavity finesse scan using a different cavity fiber mirror achieving a higher finesse on a slightly different position on the membrane. (f) Measured cavity finesse along the dashed blue line in (f). The red line shows a fit.

of the probe laser used for the finesse measurements. Here, a steep slope corresponds to a mode with high air-like character and a flat slope to a mode with high diamond-like character<sup>19</sup>. We define the mode character by assigning a value of 1 for the steepest slope (air-like, A) and 0 for the lowest slope (diamond-like, D) with corresponding values in between (see supplementary material).

The results are shown together with a corresponding measurement of the finesse in figure 3 (c). The mode character follows closely the finesse and the transmission of the cavity, proving that the losses of the cavity are dominated by effects associated with the character of the hybridized mode. To evidence the strong correlation between the cavity losses and the mode composition again over a larger region of the membrane, we scan the fiber similar to the lateral transmission measurements across the membrane and measure the finesse of the cavity at each position. The resulting finesse scan in figure 3 (d) confirms the correlation between cavity finesse and mode composition obtained along the single line described above.

We repeat the measurement with a second fiber mirror  $F_B$  since we observed high losses for the first fiber mirror already without diamond membrane. Taken at a slightly different position, the measurement shown in figure 3 (e) again reflects the shape of the membrane. The finesse spans values between  $\sim 3000$  and slightly above 20,000, which remains lower than the value observed for the cavity without membrane  $\mathcal{F}_{\max}^{\text{bare}} = 32,400$  (see supplementary material). To understand the contribution of different sources of loss, we analyze a line cut through the data shown in figure 3 (e) and fit

it with a model containing mode-dependent mirror transmission, scattering losses at the diamond-air interface, and absorption from the diamond<sup>29</sup>, see figure 3 (f). Therefore, we describe the finesse  $\mathcal{F} = 2\pi/\mathcal{L}_{\text{eff}}$  with the effective losses<sup>29</sup>

$$\mathcal{L}_{\text{eff}} = \frac{E_{\max,a}^2}{n_d E_{\max,d}^2} \mathcal{L}^f + \mathcal{L}^m + \mathcal{L}_{\text{scat}}(\sigma_{\text{rms}}) + \mathcal{L}_{\text{abs}}(\alpha_d) \quad (1)$$

Here,  $\mathcal{L}^m$  ( $\mathcal{L}^f$ ) are the losses due to transmission at the planar (fiber) mirror that we extract by a transfer matrix model using the measured DBR-layer thicknesses from the manufacturer. The scattering loss

$$\mathcal{L}_{\text{scat}} = \sin^2\left(\frac{2\pi n_d t_d}{\lambda_0}\right) \cdot \frac{(1+n_d)(1-n_d)^2}{n_d} \cdot \left(\frac{4\pi\sigma_{\text{rms}}}{\lambda_0}\right)^2, \quad (2)$$

originates from the diamond surface roughness  $\sigma_{\text{rms}}$ . In addition, we include absorption loss  $\mathcal{L}_{\text{abs}} \approx 2\alpha_d t_d$  described by the absorption coefficient  $\alpha_d$  of the diamond sample. For the general grade diamond sample, which contains an increased nitrogen concentration  $> 100$ ppb, a broadband absorption with  $\alpha_d \sim 0.1 - 0.5 \text{ cm}^{-1}$  is expected<sup>38</sup>.

For all loss contributions, knowledge of the diamond thickness and the local mode character are important. Therefore, we evaluate a measurement of the cavity mode dispersion at one location, i.e., the mode frequency shift as a function of the mirror separation, and fit a simulation to the measured dispersion to obtain the thickness  $t_{d,0} = 5.96 \mu\text{m}$ . For the other locations along the line, we make use of the known mode composition change from diamond-like (D) to air-like (A) between a local finesse minimum and maximum, corresponding to a

thickness change of  $\lambda/(4n_d) \approx 66$  nm. We interpolate the membrane thickness along the line by a linear increase between consecutive extrema.

We then perform a fit<sup>39</sup> based on the described model and the interpolated thicknesses for the finesse data along the blue line in figure 3 (e) with the surface roughness  $\sigma_{\text{rms}}$  and the absorption coefficient  $\alpha_d$  as free parameters. The result is plotted in figure 3 (f) together with the measured data. We note that the deviation between fit and data around pixel 75 originates from a local defect present on the membrane, which can be seen at the respective position in figure 3 (e). From the fit we obtain a roughness of  $\sigma_{\text{rms}} = (1.02 \pm 0.03)$  nm and an absorption coefficient of  $\alpha_d = (0.19 \pm 0.01)$  cm<sup>-1</sup>. While the absorption coefficient lies within the expected range, the predicted surface roughness is significantly higher than the measured values of  $\sigma_{\text{rms}} \approx 0.4 - 0.5$  nm<sup>23</sup>. Therefore, we conclude that we still miss further loss contributions. Possible origins can be scattering at the second diamond interface facing the mirror, surface absorption due to sp<sup>2</sup> carbon and surface defect states, and loss originating from the mismatch between the curved cavity mode phase front and the planar membrane surface (see below).

## B. Increased cavity losses due to transverse mode mixing

In most spatial maps of the diamond membrane, we observe sharp lines of constant mode character where the cavity finesse and transmission are significantly lower than in the surrounding. Similar but less severe behavior has been observed also for bare mirrors, and resonant transverse-mode mixing has been identified as the origin<sup>34,35</sup>. It occurs when higher-order transverse modes of a neighboring longitudinal mode order  $q - 1$  become resonant with a fundamental cavity mode of order  $q$ , and when boundaries such as the mirror surfaces do not match the phase fronts of Hermite-Gaussian modes, such that scattering leads to a coupling and hybridization of (near-) resonant modes. The effect is strongly dependent on the cavity length, and at large mirror separation, where low-order transverse modes fulfill the mode-mixing resonance condition, it becomes more dominant. To analyze the effect in the presence of a diamond membrane, we record transmission maps of one area (similar to figure 3 (a)) and change the air gap of the cavity over a large range. Six different scans are shown in figure 4 (a). As we increase the effective cavity length, we observe an overall transmission decrease as well as the sharp mode-mixing lines which become increasingly prominent. Notably, also at shortest mirror separation, we always observe mode-mixing lines in diamond-like regions, while they only appear at large distance in air-like regions.

To see whether the overall decrease in transmission with increasing mirror separation correlates with the cavity finesse, we perform finesse scans on an air-like sub-region for three different cavity lengths, see figure 4 (b). We observe no significant drop of the maximum finesse for the different cavity lengths, and only a weak sign of mode mixing within the studied cavity length range. This indicates that the transmission drop is dominated by mode matching.

In contrast, we have observed that losses under air-like conditions increase for thicker diamond membranes. To study this effect, we use a second diamond sample of electronic grade quality which features several membranes with different thicknesses and that was bonded on a second cavity mirror  $M_B$  (see<sup>23</sup> for details on the sample). We perform cavity transmission scans on regions of different thicknesses between 6  $\mu$ m and 19.5  $\mu$ m and measure the cavity finesse on bright (high air-like character) and dark (high diamond-like character) regions. As depicted in the central panel of figure 4 (c), the cavity finesse on bright regions shows a strong decrease with increasing membrane thickness, dropping significantly faster than expected for bulk absorption. At the same time, we observe no drop of the cavity finesse on dark regions (diamond-like conditions) with increasing membrane thickness, shown in the lower panel of figure 4 (c). We note that for the investigated effective cavity lengths, the empty cavity shows a stable finesse (see figure 4 (c), upper panel) which is valid even for longer cavity lengths (for details see supplementary material), thus ruling out effects related to the cavity fiber, i.e. clipping losses, as the origin of our observation. A possible explanation is the shape mismatch of the diamond-air interface with the curved wavefront of the cavity mode<sup>19</sup>, which becomes more prominent with increasing membrane thickness. There, the membrane thickness approaches the Rayleigh range of the cavity mode, and the curvature of the phase front at the diamond surface increases. Under such conditions, the cavity mode can no longer fulfill air-like conditions over the entire mode cross section, and an electric field at the interface will be present at the outer parts of the mode. This effectively reduces the air-like character of the mode and thus leads to increased scattering loss and mode mixing. As shown in the central panel of figure 4 (c), we can model the behavior with a simple estimate where we calculate the diamond / air character based on the weighted fraction of electric field present at the diamond-air interface due to the wavefront curvature for a given cavity and membrane geometry (details on the model can be found in the supplementary material). Consistent with this picture, we observe no decrease of the cavity finesse under diamond-like conditions for increasing membrane thickness.

Beyond this general trend which is present on the entire membrane and for any air gap, our measurements show that the diamond leads to significant resonant coupling between different transverse modes. To directly reveal the mode coupling, we use the cavity with fiber mirror  $F_B$  and the general grade sample to scan laterally in fine steps over a region where the transmission drops sharply and record the cavity modes at each position. We show transmission maps on three different areas where mode coupling occurs in figure 5 (a,b,c). To display the coupling of higher-order modes to the fundamental mode, we record cavity transmission spectra taken by varying the cavity length, for each position along three different linear paths indicated in figure 5 (a,b,c), see figure 5 (d,e,f), respectively. At locations where a fundamental mode and a higher-order mode would show degeneracy, avoided crossings appear, and the transmission drops. Figure 5 (d) shows the cavity spectra for a scan across an air-like region. The first spectrum (red, bottom) features one prominent fundamental

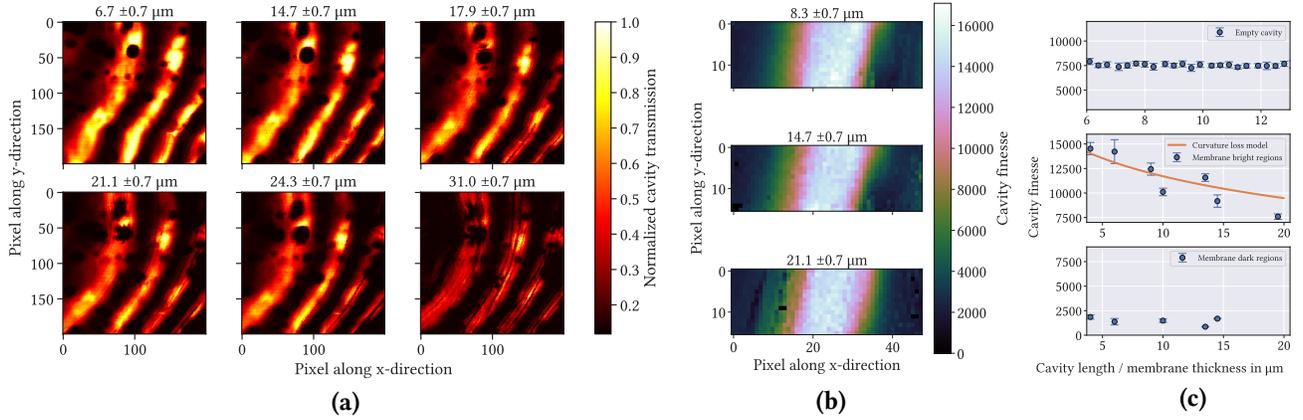


FIG. 4. (a) Cavity transmission scans for increasing air gap and fixed membrane thickness. The effective cavity length is given above each scan. The scans are normalized to the maximum value of all scans. (b) Cavity finesse scans for different effective cavity length by changing the air-gap on an air-like region. (c) Cavity finesse of the empty cavity (upper panel) for different cavity length and finesse on bright regions (central panel) and dark regions (lower panel) on the membrane for different membrane thicknesses. Each data point shows the average of at least 100 measurements and the orange curve in the central panel shows a model based on additional losses for air-like modes coming from the mode curvature at the diamond-air interface. For easier comparison, the range of shown cavity lengths in the upper panel corresponds to the effective cavity lengths resulting from the membrane thickness and the air gap of the lower two panels.

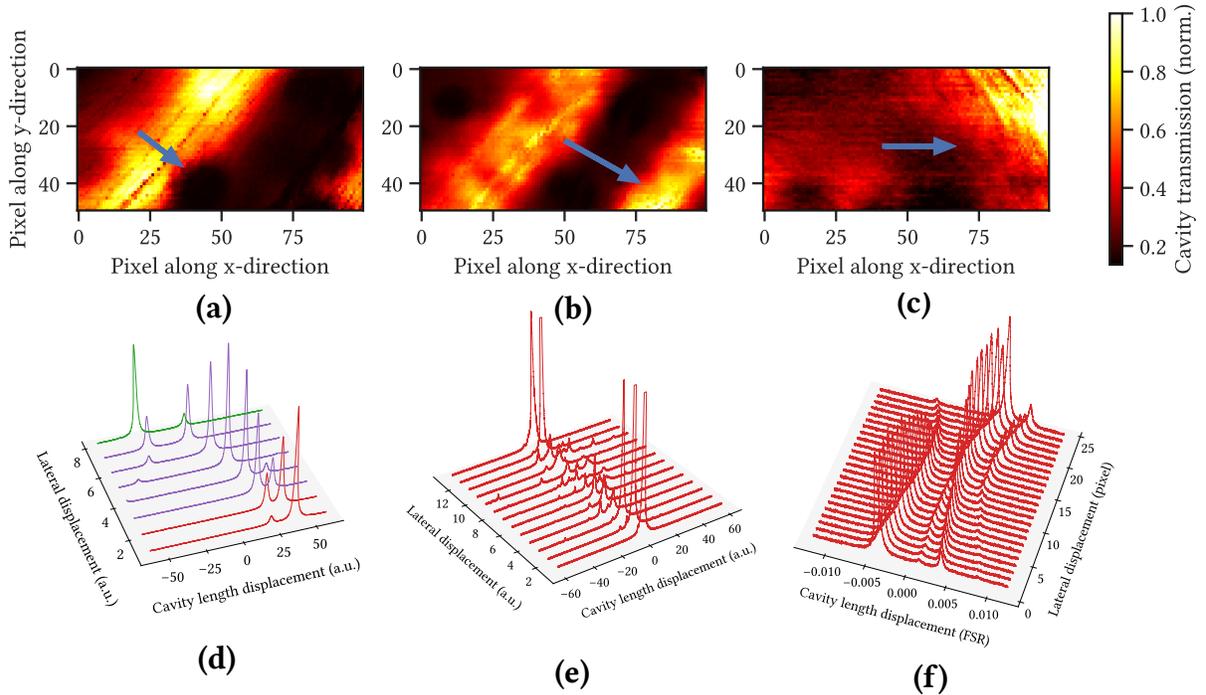


FIG. 5. (a)-(c) Cavity transmission scans of areas with resonant mode coupling. Blue arrows indicate the location and direction of the scans shown in (d) - (f). (d) Cavity transmission spectra under air-like conditions for positions along the blue arrow in (a). The color is changed after each avoided crossing for better visibility. (e) Transmission spectra under diamond-like conditions along the arrow shown in (b). Coupling between the fundamental mode and a series of higher-order modes leads to a strong decrease of the cavity transmission. (f) Cavity transmission spectra for positions along the arrow shown in (c).

mode and a second, higher-order mode approaching from the left. When both modes would become degenerate, the coupling leads to an avoided crossing. In this example, a second avoided crossing with another transverse mode and larger coupling is visible at small spatial distance.

Under diamond-like conditions, mode mixing is observed to be more severe, and it can include a large number of consecutive avoided crossings such that the transmission drops almost completely, see figure 5 (e). Here, the laser power is increased to saturate the APD outside the avoided crossings

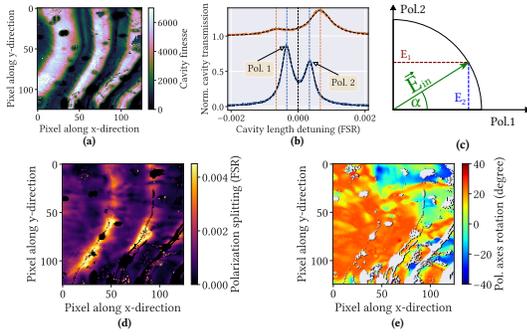


FIG. 6. Polarization splitting and rotation of the fundamental cavity modes. **(a)** Cavity finesse scan on the diamond membrane. **(b)** Fundamental cavity modes on a position with high (blue) and low (orange) finesse. The data for low finesse is shifted along the y-axis for better visibility. Black dashed lines are Lorentzian fits on the transmission data. Vertical lines show the different amount of mode splitting. **(c)** The intensity ratio of the polarization modes can be used to determine the relative polarization angle between incoupled light and the cavity polarization modes. **(d)** Lateral scan of the polarization splitting in units of FSR. One finds a larger splitting for modes with higher diamond-like character. **(e)** Spatial map of the polarization angle obtained from the same data used for **(d)**.

such that the modes can still be detected while mixing. A further example with a large spatial region of mode mixing is shown in figure 5 (f). Here, one can observe the coupling of both polarization modes. For calibration of the cavity length changes, each spectrum is recorded over more than one FSR.

The alignment of mode mixing resonance contours with the diamond membrane topography can be understood by the different effect of the membrane on the frequency of different transverse modes<sup>40</sup>. Due to the larger Gouy phase of higher-order modes compared to the fundamental mode, their diamond- and air-like mode character occur for different membrane thicknesses, such that thickness variations will lead to a large variation of mode frequency differences, and transverse-mode resonance conditions will follow isocontours of constant membrane thickness.

Several contributions add to the mode mixing, such as imperfect mirror shape and misalignment of the cavity (see e.g. fig. 3 (d) vs (e)), which can be both minimized. The origin of the membrane-induced mode mixing is due to imperfect surface topography such as a wedge, but also due to the shape mismatch between the planar diamond-air interface and the parabolic wave front of the cavity mode<sup>19</sup>. Due to this curvature, an exact air-like condition with zero field at the entire interface cannot be achieved. This introduces interface-induced mode mixing also for air-like modes. For a plano-concave cavity geometry, the wave front curvature approaches a planar shape close to the waist on the plane mirror, such that for thin membranes, one can largely avoid diamond-induced mode mixing<sup>25,30</sup>.

### C. Polarization mode splitting and birefringence

As a last aspect, we investigate how the diamond membrane affects the polarization modes. For cavity mirrors with broken rotational symmetry, an intrinsic polarization mode splitting is present<sup>41</sup>, which is also the case for the cavities used here. For the empty cavity, we measure a mode splitting of  $\delta = v_{\text{split}}/v_{\text{FSR}} = 3.82 \times 10^{-5}$ , which is consistent with the measured ellipticity of the concave profile of the fiber (see supplementary material). On the diamond membrane, we observe a significantly increased and spatially varying mode splitting spanning values between  $\delta = 5 \times 10^{-4}$  and  $5 \times 10^{-3}$ . Figure 6 (a) shows a finesse scan of the investigated region of the membrane for reference of the mode character. From cavity transmission spectra on two locations with different mode character, we find a significant difference in the polarization splitting, see figure 6 (b). When measuring a map of the polarization splitting on the same area as shown in figure 6 (a), we observe fringes with the same shape as for the cavity finesse, see figure 6 (d). Positions with low finesse, i.e. diamond-like character, show a large splitting, and positions with air-like character show a smaller splitting. The increased splitting under diamond-like conditions indicates a strong contribution of the diamond-air interface to the polarization splitting. At this point, we have no model that can explain the effect.

Also for air-like modes, where the interface is "hidden" from the cavity field, we observe an increased splitting compared to an empty cavity, with a small spatial variation on a larger scale. We attribute this to birefringence of the diamond which originates from local strain. The observed diamond-induced mode splitting in air-like regions of  $\delta = 0.001$  corresponds to a refractive index difference of  $\Delta n = \lambda/t_d \cdot \delta = 1 \times 10^{-4}$ , indicating an elevated local strain level. In contrast, carefully grown diamonds can achieve one to two orders of magnitude smaller strain levels<sup>42</sup>.

Further, we observe a spatial variation of the intensity ratio of the polarization modes, originating from a rotation of the cavity polarization axes. To study this observation in more detail, we measure the intensity ratio of both polarization modes and calculate the corresponding polarization angle between the probe laser and one of the cavity modes by  $\tan(\alpha) = E_2/E_1 = \sqrt{I_2/I_1}$ , as sketched in figure 6 (c). Here  $I_1$  and  $I_2$  are the transmitted intensities of the two polarization eigenmodes. As shown in figure 6 (e), the extracted angle changes significantly over the region, showing a completely different pattern than the topography of the membrane. We ascribe this signal to the variation of the orientation of the slow axis of the birefringent diamond, which is consistent with a spatially varying strain. To exclude measurement artefacts, we have repeated the measurements with a second cavity with the fiber mirror  $F_B$  at a slightly shifted position (see supplementary material), and observe the same pattern. For comparison, we also studied a second electronic grade diamond sample in a cavity. There, we observed mode-character dependent polarization splitting between  $\delta = 2 \times 10^{-4}$  and  $9 \times 10^{-4}$ , resulting in a reduced birefringence of  $\Delta n \approx 2.5 \times 10^{-5}$  (see supplementary material). This lower level of birefringence together with the splitting induced by the ellipticity of the fiber mirror can

lead to a polarization mode splitting that remains smaller than a cavity linewidth as long as the finesse remains smaller than  $\approx 5000$ .

It remains an interesting question whether noticeable strain can originate from the van der Waals bond, e.g. for non-perfectly planar interfaces. This contribution could be quantified by measuring the birefringence before and after bonding. Birefringence can become a limiting factor for aligning the cavity mode polarization with the transition dipole orientation of color centers. This can reduce the achievable coupling strength, lead to issues with circularly polarized transitions, and limit the performance of the crossed polarization detection scheme<sup>24</sup>. On the other hand, suitable polarization mode splitting can be made use of for polarization-selective Purcell enhancement for efficient resonance fluorescence collection<sup>43,44</sup>.

#### IV. CONCLUSIONS

In our study we have used scanning cavity microscopy to evidence the effects of the topography of a diamond membrane on the cavity modes. The diamond- and air-like character strongly dominates the mode properties and cavity loss, and we were able to differentiate the different loss contributions and show the importance of absorption loss for samples with increased nitrogen concentration. We have further identified transverse mode mixing as a limiting mechanism when operating under diamond-like conditions, and loss of purely air-like conditions for thick membranes. These limitations are expected to be avoided by using thinner membranes. Finally, we have introduced a technique to sensitively measure local birefringence and observed a mode-character dependent polarization mode splitting and spatial variation of diamond birefringence. Scanning cavity microscopy thereby provides important insight into the required properties of diamond membranes for optimized spin-photon interfaces.

#### SUPPLEMENTARY MATERIAL

See supplementary material for details on the cavity fibers and mirrors, calibration of the nanopositioning system, evaluation of the hybridized mode composition, measurements of the rotation of the polarization axes and on the second diamond sample of electronic grade quality.

#### AUTHOR DECLARATIONS

##### Conflict of Interest

The authors have no conflicts to disclose.

#### Author Contributions

**Jonathan Körber:** Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Software; Validation; Visualization; Writing - original draft; Writing - review & editing. **Maximilian Pallmann:** Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Validation; Visualization; Writing - original draft; Writing - review & editing. **Julia Heupel:** Data curation; Investigation; Resources; Writing - review & editing. **Rainer Stöhr:** Resources; Writing - review & editing. **Evgenij Vasilenko:** Investigation; Writing - review & editing. **Thomas Hümmer:** Resources; Software; Writing - review & editing. **Larissa Kohler:** Investigation; Resources; Writing - review & editing. **Cyril Popov:** Conceptualization; Funding acquisition; Resources; Supervision; Writing - review & editing. **David Hunger:** Conceptualization; Funding acquisition; Methodology; Project administration; Supervision; Validation; Writing - original draft; Writing - review & editing.

#### DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

#### ACKNOWLEDGMENTS

We acknowledge experimental support from Jonas Grammel, Timon Eichhorn, Tobias Krom, and Julia Benedikter. This project received funding from the European Union Horizon 2020 research and innovation program within the Quantum Flagship project SQUARE (Grant Agreement No. 820391), the German Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung, BMBF) within the project Q.Link.X (Contracts No. 16KIS0879 and 16KIS0877), QR.X (Contracts No. 16KISQ004 and 16KISQ005), SPINNING (Contract No. 13N16211), and NEQSiS (Contract No. 16KISQ029K), and the Karlsruhe School of Optics and Photonics (KSOP).

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