

Maser threshold characterization by resonator Q-factor tuning

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Whereas the laser is nowadays an ubiquitous technology, applications for its microwave analog, the maser, remain highly specialized, despite the excellent low-noise microwave amplification properties. The widespread application of masers is typically limited by the need of cryogenic temperatures. The recent realization of a continuous-wave room-temperature maser, using NV^- centers in diamond, is a first step towards establishing the maser as a potential platform for microwave research and development, yet its design is far from optimal. Here, we design and construct an optimized setup able to characterize the operating space of a maser using NV^- centers. We focus on the interplay of two key parameters for emission of microwave photons: the quality factor of the microwave resonator and the degree of spin level-inversion. We characterize the performance of the maser as a function of these two parameters, identifying the parameter space of operation and highlighting the requirements for maximal continuous microwave emission.

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The first maser system was realized using ammonia molecules in the gas phase¹ and applications for signal amplifiers, frequency standards or as spectrometers were subsequently proposed. However, the need for cryogenic and/or high vacuum environments restricted miniaturization and integration towards more general applications. Maser research and development was mainly focused on low-noise microwave receiving systems for deep-space antenna networks², and several other maser systems based on ruby³, atomic hydrogen⁴ or Rydberg atoms⁵ were realized. These systems were all still subject to the same restrictions and the focus on fundamental research into masers declined. The field was reinvigorated upon the realization of a room-temperature pulsed maser in an optically pumped crystal of pentacene-doped *p*-terphenyl, placed inside a high quality factor microwave resonator^{6–8}. Here, masing was achieved not only without the need of cryogenics or a high-vacuum environment, but also with easily accessible optical pump rates. Shortly afterwards, a proposal for a continuous-wave room-temperature maser in optically spin polarized, negatively charged nitrogen vacancy (NV⁻) centers in diamond⁹ and its consecutive experimental realization¹⁰ followed. The excellent low-noise amplification properties of the maser have been demonstrated in recent work^{11–13} and has found application in enhanced quantum sensing of molecular spin ensembles¹⁴. The search for other solid-state maser materials, such as SiC¹⁵ has continued, and an application in quantum technology by creating a maser system based on Floquet states in Xe atoms¹⁶ has also been reported.

With the increased focus on room-temperature solid-state masers a quantitative experimental characterization of the parameter space of operation supplements the current endeavors to optimize the performance of such systems. Understanding the behavior of the maser performance, defined by the level-inversion of the spin ensemble and the loaded Q -factor Q_L of the resonator, can lead to higher output power maser systems. To guarantee reproducibility and maximize the power output of maser-based technologies, a complete understanding of the system parameter space, including the minimal requirements to surpass the masing threshold, is required.

Here, we present an experimental setup that we exploited to investigate the maser performance of a NV⁻ spin ensemble hosted in diamond as a function of the resonator quality factor and the degree of spin level-inversion. In the resulting maser threshold diagram, we can clearly identify the threshold for maser action, thereby obtaining a set of experimental boundary conditions for the optimal operation of a maser system. Additionally, our optimized setup yields the highest continuous-wave maser output power reported to date.

Results and discussion

Experimental setup. We use a cylindrical dielectric ring resonator made of sapphire to deliver/detect resonant microwaves to/from the NV⁻ centers contained in the diamond host. A key parameter for the continuous maser emission is the loaded Q -factor, Q_L , of the resonator, which is defined by $1/Q_L = 1/Q_{\text{int}} + 1/Q_{\text{ext}}$ with the internal Q -factor, Q_{int} , and the external Q -factor, Q_{ext} . Although sapphire dielectric resonators exhibit low dielectric losses¹⁷, radiative losses typically dominate and prevent high internal quality factors. Hence, to suppress radiative losses the resonator is placed inside a metal cavity. The cavity design is further constrained by two conditions: (i) it has to fit between the poles of our electromagnet system, which gives the static magnetic field B_0 used to tune the energy levels of the NV⁻ spins via the Zeeman interaction, and (ii) the resonance frequency of the cavity containing the sapphire ring is required to be within the

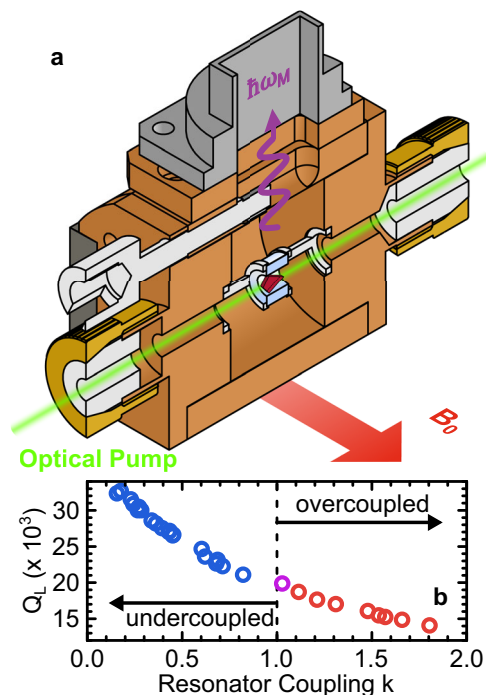


Fig. 1 Schematic cross-section of the microwave cavity-resonator setup.

a Microwave cavity, holding the sapphire ring resonator. The schematic shows half of the cavity to reveal the interior. The light blue sapphire ring is held by two Teflon holders. On the top is the iris coupled waveguide port. The coupling to the resonator is adjusted by a Teflon screw with a metal ring at its tip. The 532 nm optical pump laser is aligned along the symmetry axis of the cylindrical resonator, while the static magnetic field B_0 is perpendicular to it. Finally, the diamond sample is placed inside the resonator. **b** Loaded Q -factor as a function of the resonator coupling, showing the full range of coupling, with the diamond sample in the resonator.

9–10 GHz (X-band) frequency range, to allow a fast pre-characterization of the NV⁻ spin ensemble by conventional electron spin resonance (ESR). To this end, we designed a cylindrical cavity made of oxygen-free high-thermal conductivity copper, plated with thin layers of (first) silver and (second) gold to prevent oxidation of the metal surfaces, thereby minimizing additional resistive losses.

Figure 1a shows a 3D schematic of the fully assembled cavity cut at a symmetry plane to reveal the interior. The cavity has sample entries at opposite ends of the cylinder symmetry axis for sample and optical access. In the center of the cavity the resonator is held in place by two wire-frame Teflon holders. The holders are machined to have minimal volume in order to minimize additional dielectric losses. Microwave power is coupled in and out via a single waveguide iris port on the top. This port represents the external quality factor Q_{ext} and together with Q_{int} the resonator coupling $k = Q_{\text{int}}/Q_{\text{ext}}$ is defined. k is controlled via a Teflon screw with a metal ring at its tip. Changing the coverage of the iris by the metal ring allows a continuous change¹⁸ from over-coupled to under-coupled: the regimes where Q_L is dominated by external losses or intrinsic losses, respectively. Figure 1b shows the achievable Q_L as a function of k for our resonator-cavity system, loaded with the diamond sample. The resonator parameters are extracted from microwave reflection measurements, using a vector network analyzer. By fitting the microwave reflection as a function of frequency with a Lorentzian model function the parameters for resonance frequency, resonator coupling and internal quality factor are determined. A

description of the model function can be found in the methods section. We use the $TE_{01\delta}$ mode for our experiments, where the electric field is mostly contained in the sapphire ring and the magnetic field is mostly focused in the bore. Without a sample, the sapphire resonator has a resonance frequency $\omega_{\text{res}}/2\pi = 9.25$ GHz and the $Q_L = 42,500$ when fully under-coupled. We define the resonator as fully under-coupled when $k = 0.0027$, where the Iris is no longer covered by the metal tip of the Teflon screw. Here, Q_L remains unchanged upon further extraction of the screw. For detailed cavity and resonator dimensions see Supplementary Note 4¹⁹.

Maser working principles. The process of continuous emission of microwave photons from the NV^- centers is schematically shown in Fig. 2a, c. The diamond hosting the NV^- centers is placed inside a high-Q resonator, which is highly under-coupled ($k \ll 1$). The resonator may be pictured in analog to its optical counterpart the laser, with one perfectly reflective mirror and one weakly transmitting mirror. The latter represents the iris coupled single microwave port on the microwave cavity.

The applied static magnetic field B_0 lifts the degeneracy of the $|\pm 1\rangle$ states and, tunes the $|-1\rangle$ state energetically below the $|0\rangle$ state such that the splitting $\hbar\omega_M$ is resonant with the microwave resonator frequency. The experiment is performed only on one sub-set of NV^- centers which are aligned with the external magnetic field B_0 . This provides the shown level structure, having the largest Zeeman splitting of the energy levels and consequently the largest initial population difference at Boltzmann equilibrium. By illuminating the NV^- centers continuously with a 532 nm laser, the spin populations which are initially at Boltzmann equilibrium are predominately pumped into the $|0\rangle$ state²⁰, resulting in a level-inversion (see Fig. 2a). A description of the optical spin polarization process is found in Supplementary

Note 2¹⁹. Finally, the laser polarization is required to be aligned along the NV^- defect axis to achieve most efficient pump rates^{21,22}.

To trigger a collective stimulated emission, an initial photon with $\hbar\omega_M$ is required. This is provided either by an externally applied seeding photon, due to spontaneous emission or thermal photons (see Fig. 2b). From this point an avalanche of stimulated photons is created, forming a coherent microwave field inside the resonator (see Fig. 2c). If the laser pump rate is sufficient to maintain the level inversion and the resonator loaded quality factor is high enough to support a large enough coherent microwave field, continuous microwave emission is achieved.

NV defect axis alignment. In our experimental setup, the diamond sample is held inside a quartz ESR tube, supported between two additional quartz tubes fitted inside the first one. The tube is inserted into the cavity and positioned such that the diamond is located at the center of the sapphire ring where the magnetic component of the microwave field is largest. The cavity with sample is mounted between the poles of a electromagnet system. We connect either a conventional ESR spectrometer, a vector network analyzer or a spectrum analyzer to the microwave port of the cavity, to perform low-power microwave spectroscopy of the NV^- spin transitions or to study the maser emission. A goniometer is attached to the sample tube, allowing a precise rotation of the quartz tube containing the diamond with respect to the static magnetic field. Conventional ESR as a function of B_0 and rotation angle is performed to find an orientation of the diamond where the defect axis of a sub-set of NV^- centers is mostly parallel to the applied magnetic field. For such an orientation the NV^- spins feature an energy level scheme of the electronic ground state as schematically shown in Fig. 2a and the states $|0\rangle$ and $|\pm 1\rangle$ can be considered pure. This can be characterized by the frequency/magnetic field splitting between the low-field ($|0\rangle \mapsto |+1\rangle$) and high-field ($|0\rangle \mapsto |-1\rangle$) transitions corresponding to twice the zero-field splitting D . Away from this alignment the states become mixed, resulting in a smaller splitting than $2D$ between the two allowed transitions^{23,24} and a smaller maximal achievable spin polarization. Without laser illumination, we find a maximal splitting of about 205 mT or 5.762 GHz, agreeing well with twice the zero-field splitting, D , of NV^- centers in diamond.

Maser emission spectrum characterization. Having optimized the orientation of the NV^- centers, we characterize the performance of our maser setup by analyzing the microwave emission spectrum. Figure 3a shows the color encoded maser emission power $|A_M|$ as a function of frequency and static magnetic field for a laser pump rate $w_L = 430$ s⁻¹ and with the resonator fully under-coupled, $Q_L \approx Q_{\text{int}}$, which gives a loaded quality factor Q_L of 33,500. The three bright lines represent the maser emission of the three ¹⁴N hyperfine transitions of the NV^- centers oriented along B_0 . In this configuration, we achieve a maximum maser emission power of -56.5 dBm. The maser emission shows a finite frequency-magnetic field dispersion, where the maser power is maximal in the center of the line. This is illustrated in Fig. 3b which depicts the maser emission power as a function of frequency for two fixed magnetic fields. The dispersion results from the hybridization of the microwave resonator mode and the resonant NV^- transition. The resonator resonance frequency $\omega_{\text{res}}/2\pi = 9.12$ GHz lies at the center of the middle maser emission line. The inverted spin population causes a dispersive shift of the resonator frequency to lower frequencies for the lower magnetic field emission line and respectively to a shift to higher frequencies for the higher magnetic field emission line. The

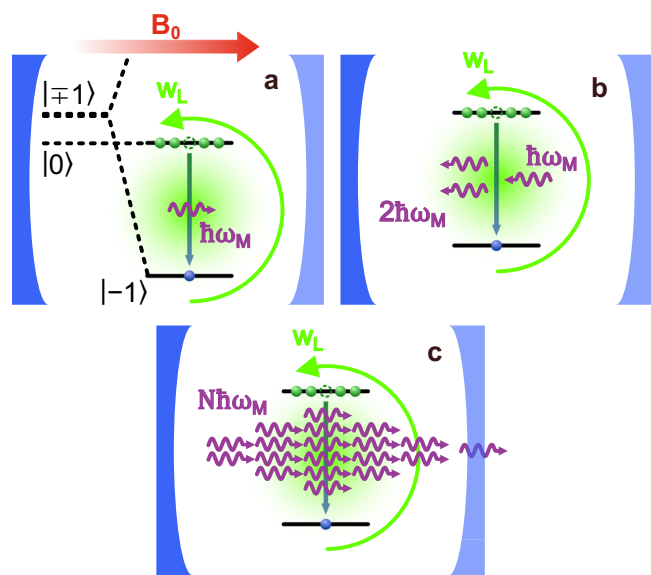


Fig. 2 Schematic of the maser emission process. a-c The blue mirrors are the optical analog of a high-Q microwave resonator, with one mirror being fully reflective (dark blue) and one weakly transmissive (light blue); **(a)** Energy states of the NV^- spin ensemble aligned along B_0 . The $|-1\rangle$ state is tuned below the $|0\rangle$ state by the Zeeman interaction, while the optical pump generates a population inversion and spins spontaneously emit microwaves. **b** Spontaneously emitted or thermally stimulated microwaves can stimulate the emission of additional microwaves. **c** For sufficiently large optical pump rates and a high Q_L coherent microwave emission is established.

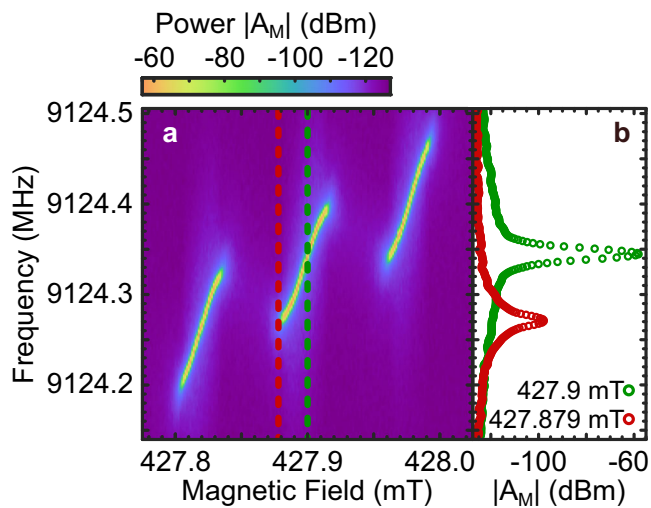


Fig. 3 Maser emission spectrum. **a** Maser emission power $|A_M|$, in logarithmic units, as a function of frequency and static magnetic field at $w_L = 430 \text{ s}^{-1}$ and Q_L of about 33,500. **b** $|A_M|$ as a function of frequency for two different magnetic field values, indicated by the red and green dashed lines in (a).

maser dispersion can be described via a Tavis-Cummings model with an inverted spin polarization, where the frequency range covered by the maser emission lines increases with increasing inversion.

In order to determine the dependency of the maser output on the laser pump rate w_L and the loaded quality factor Q_L , both were varied systematically. The latter is dependent on the resonator coupling k which is controlled by the iris screw. Figure 4a shows the peak maser power $|A_M|$ of the central maser line as a function of w_L (from 165 s^{-1} to 695 s^{-1}) and Q_L (from 14,000 to 33,500). Note, that after each change of w_L we wait 45 min before starting measurements to allow the diamond to reach a thermal equilibrium (see Supplementary Note 1 for details¹⁹). For low loaded quality factors, i.e., an over-coupled resonator, there is no microwave emission for all laser pump rates studied and the signal amplitude is represented by the noise floor of the spectrum analyzer. Note that critical resonator coupling ($k = 1$) is achieved at $Q_L \approx 20,000$, marking the transition between over- and under-coupling²⁵. With increasing Q_L a weak microwave emission is observed for moderate to high laser pump rates. Here, the emission spectrum is broad and governed by the amplification of thermal photons residing in the resonator⁹. In this region, the resonator losses are still too high to allow the build-up of sufficient stimulated photons for continuous masing. For $Q_L > 26,000$ and $w_L > 400 \text{ s}^{-1}$ the rate of stimulated emission exceeds the losses in the resonator and the spin system, and continuous masing is established. Figure 4b shows the evolution of the peak maser power as a function of the loaded quality factor for two different laser pump rates. Upon passing the threshold Q_L of 26,000, $|A_M|$ increases until reaching an onset of saturation for the highest Q_L , where the final level of saturation depends on the laser pump rate. A Q_L larger than 26,000 further promotes the stimulated microwave emission, thus reducing the threshold laser pump rate required. For a fully under-coupled resonator with $Q_L = 33,500$, continuous microwave emission is already achieved for $w_L > 200 \text{ s}^{-1}$. Figure 4c shows the peak maser power $|A_M|$ as a function of the laser pump rate w_L at two different loaded quality factors of the resonator. The peak maser power increases with increasing laser pumping, reflecting the achieved level-inversion. Consequently, for the largest Q_L and highest w_L , which

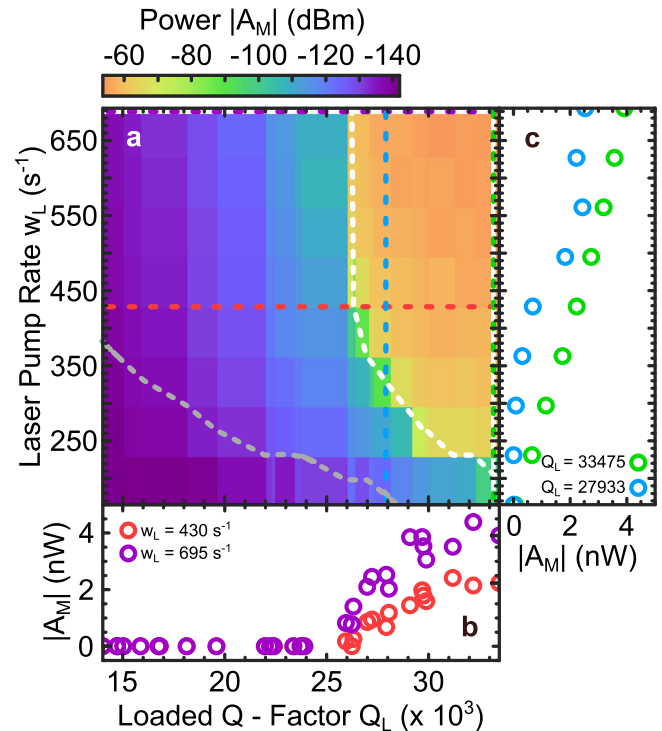


Fig. 4 Maser threshold diagram. **a** Maximal maser emission power $|A_M|$, in logarithmic units, of the central hyperfine maser transition line as a function of laser pump rate w_L and the loaded quality factor Q_L of the resonator. The white and gray dashed lines represent the theoretical threshold for masing, with and without temperature effects, respectively. **b** $|A_M|$ as a function of Q_L for two w_L , indicated by the purple and red dashed lines in (a). **c** $|A_M|$ as a function of w_L at two Q_L , indicated by the blue and green dashed lines in (a).

guarantees a stable microwave field inside the resonator and the largest level inversion, respectively, the highest maser power of -54.1 dBm is achieved. This is an improvement of more than three orders of magnitude in maser power over the initial report on a NV⁻-based maser in ref. ¹⁰ with reported -90.3 dBm . The threshold laser pump rate w_{th} for masing can be derived from the steady-state Heisberg equations of motion based on the coupled resonator-spin ensemble Tavis-Cummings model, including dissipative channels for the resonator and spins, and is given by^{9,10,26}

$$w_{th} = \frac{\eta}{T_1 \left(\frac{g_0^2 N}{\kappa_0 \gamma} - 1 \right)}. \quad (1)$$

Here, T_1 is the effective longitudinal relaxation time of the NV⁻ spins, g_0 gives the strength of the magnetic dipole coupling between a single spin and a single microwave photon²⁷, N is the absolute number of spins per hyperfine transition and per NV⁻ defect axis and κ_0 and γ are the resonator and spin loss rates (HWHM), respectively. We estimate $g_0/2\pi$ by finite element simulations of the magnetic field profile inside the sapphire ring resonator²⁸ to an average value of 0.244 Hz and determine $\gamma/2\pi$ and N via low-power microwave spectroscopy of the low-field transition ($|0\rangle \leftrightarrow |+1\rangle$) to $530.8 \text{ kHz} \pm 7.7 \text{ kHz}$ and 2.32×10^{13} , respectively. A detailed derivation of these system parameters is found in Supplementary Note 3¹⁹. The resonator loss rate is defined through Q_L as $\kappa_0 = \omega_{res}/2Q_L$, which we control by changing the resonator coupling. The scaling factor η modifies the optical pump rate defined for a two-level system within the framework of the Tavis-Cummings model to take all seven energy levels involved in the pumping of a single NV⁻ spin into account.

We extract $\eta = 14.05$ from the calculated spin-level inversion as a function of $w_L^{9,10}$, by solving the set of optical pump rate equations in the steady-state²⁹. For T_1 we explicitly take the influence of an optical pump into account. In addition to polarizing spins, the pump leads to excessive heating of the diamond, which decreases the T_1 time³⁰. We determine T_1 for each w_L , finding 5.2 ms for low w_L and a minimum of 1.5 ms for the highest w_L , where a detailed description of the T_1 dependence on w_L is given in Supplementary Note 1¹⁹. The white dashed line in Fig. 4a gives the masing threshold w_{th} as a function of the loaded quality factor and is in excellent agreement with the experimentally found threshold of our maser, when acknowledging the reduction of the relaxation time due to high pump rates. For comparison, we include the expected threshold for a fixed $T_1 = 5.2$ ms (gray dashed line), demonstrating the significant influence of laser heating.

Conclusion

To summarize, our characterization setup allows the resonator coupling to be continuously and precisely adjusted between over- and under-coupled in conjunction with the optical pump rate. This permits the detailed study of the performance of the maser as a function of the rate of stimulated emission and the degree of level-inversion. Control of these parameters enabled the first experimental verification of the maser threshold equation over a wide parameter space. Thus, the regions of microwave emission below the detection limit, thermal photon amplification and continuous masing could be identified in a NV⁻ diamond maser.

Our results highlight an efficient operation of the maser is in the highly under-coupled regime. Clearly, this limits the maser output power as only a small fraction of the microwaves in the weakly coupled resonator can exit. For applications, a higher output is essential and, hence, a larger coupling to the resonator is required. The resulting threshold diagram suggests that either higher pump rates or a resonator with a higher Q_{int} will enable a larger coupling of the resonator. However, higher pump rates not only require bulky laser systems but cause the sample to heat up. We demonstrate that the T_1 relaxation time is shortened, thus reducing the spin inversion. Indeed, it is clear for our results that pump rates can be reduced to a level where small (e.g., <1 W) laser diodes can be employed instead. Therefore, increasing the Q factor of the resonator is a more viable approach, although it is necessary to point out that this will decrease the bandwidth¹¹. Finally, the optimized design yielded an increase of the maximal maser output of more than three orders of magnitude compared to the initial report on a NV⁻-based maser from -90.3 dBm to -54.1 dBm¹⁰. We attribute this mainly to an improved heat management of the diamond sample to limit a reduction in T_1 time and consequently limit the increase of the masing threshold. Thus it provides not only a blueprint for solid-state based maser systems and but also sets a benchmark for future characterization and optimization studies.

Methods

Diamond sample. Our diamond sample is of rhombic shape with its long axis having about 5 mm, its short axis having about 4 mm and a thickness of 1 mm. The diamond consists of natural abundance carbon and we estimate the total number of NV⁻ to 2.78×10^{14} or 0.16 ppm (see Supplementary Note 3 for details on the number of spins estimate¹⁹). At room temperature and no optical pump the NV⁻ feature a $T_2 = 25$ μ s, determined by pulsed ESR.

Laser pumping. We use a 532 nm *Coherent Verdi V-5* laser to optically pump the spin population of the NV⁻ centers. The laser

features a spot size of about 4 mm and hits the diamond sample on its flat edge, an area of about 4 mm \times 1 mm. We determine the laser pump rate as $w_L = \sigma P_{pump}/A_{pump} \hbar \omega_{pump}$ (ref. 10), with the one-photon absorption cross-section for NV⁻ centers $\sigma = 3.1 \times 10^{-21}$ m², the pump laser power P_{pump} , the laser spot area A_{pump} and the laser frequency ω_{pump} .

Microwave spectroscopy. We study the microwave emission from the maser with a Keysight N9020B MXA 10 Hz to 44 GHz spectrum analyzer. To improve the SNR we pre-amplify the maser signal, using a Mini Circuits Low Noise Amplifier ZX60-06183LN + prior detection by the spectrum analyzer. The low power microwave spectroscopy was carried out using an Anritsu vector network analyzer MS46122B 1 MHz to 43.5 GHz. For conventional ESR measurements a Bruker EMXplus spectrometer is used.

Resonator coupling. We determine the microwave resonator coupling k by measuring the microwave reflection as a function of frequency with a VNA, far detuned from the NV⁻ centers. We fit the magnitude of the reflection scattering parameter $|S_{11}|$ of the resonator for different positions of the Iris screw, using a Lorentzian model function derived from an equivalent circuit model, which depends on k ³¹.

$$|S_{11}| = A \left| \frac{k - 1 - iQ_{int} \left(\frac{\omega}{\omega_{res}} - \frac{\omega_{res}}{\omega} \right)}{k + 1 + iQ_{int} \left(\frac{\omega}{\omega_{res}} - \frac{\omega_{res}}{\omega} \right)} \right|. \quad (2)$$

In addition, we can extract the internal quality factor Q_{int} and via the relation for the resonator coupling $k = Q_{int}/Q_{ext}$ the external Q -factor Q_{ext} and hence the loaded quality factor Q_L , as well as the resonator frequency ω_{res} and its loss rate κ_0 .

Data availability

The data that supports the findings of this study is available upon reasonable request.

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Author contributions

S.R., R.R. and C.W.M.K. designed and optimized the resonator. The experimental setup and its automation was realized by S.R., Y.F., H.T.A.W., C.W.Z. and C.W.M.K. C.W.Z. performed the experiments and the data analysis with input from J.D.B. and C.W.M.K. C.W.Z. supported by C.W.M.K. wrote the manuscript with input from all authors. C.W.Z., J.D.B. and C.W.M.K. conceived the study.

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Competing interests

The authors declare no competing interests.

Additional information

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