

Coupling nitrogen-vacancy center spins in diamond to a grape dimer

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Two grapes irradiated inside a microwave (MW) oven typically produce a series of sparks and can ignite a violent plasma. The underlying cause of the plasma has been attributed to the formation of morphological-dependent resonances (MDRs) in the aqueous dielectric dimers that lead to the generation of a strong evanescent MW hotspot between them. Previous experiments have focused on the electric field component of the field as the driving force behind the plasma ignition. Here we couple an ensemble of nitrogen-vacancy (N-V) spins in nanodiamonds (NDs) to the magnetic field component of the dimer MW field. We demonstrate the efficient coupling of the N-V spins to the MW magnetic field hotspot formed between the grape dimers using optically detected magnetic resonance (ODMR). The ODMR measurements are performed by coupling N-V spins in NDs to the evanescent MW fields of a copper wire. When placing a pair of grapes around the NDs and matching the ND position with the expected magnetic field hotspot, we see an enhancement in the ODMR contrast by more than a factor of 2 compared to the measurements without grapes. Using finite-element modeling, we attribute our experimental observation of the field enhancement to the MW hotspot formation between the grape dimers. The present study not only validates previous work on understanding grape-dimer resonator geometries, but it also opens up another avenue for exploring alternative MW resonator designs for quantum technologies.

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I. INTRODUCTION

Grapes are one of the most popular fruits with a lot of health benefits [1]. Since the first observation of sparks in the gap between two spatially separated grape pieces inside a microwave (MW) oven in 1994 [2], grapes became a central character in an interesting physics problem. Khat-tak *et al.* [3] provided a physical explanation for the rapid succession of sparks phenomenon by focusing on the electric field distribution around the grapes. They show that the grape dimers (or hydrogel water beads with implanted ions) act like a MW resonator where the fields are stored inside the grapes due to morphological-dependent resonances (MDRs). MDRs in grapes are caused by their curvature [4] and high-electric permittivity [e.g., $\text{Re}(\epsilon_r) \approx 80$ at 2.5 GHz [5]]. The observed sparks are due to the plasma formation from metallic ions in the grapes [6]. Motivated by this work, one might wonder whether

such grape dimer arrangements could enable technical applications where strong MW field enhancement is needed.

Microwave resonators that confine MW fields to a finite volume are employed in satellite technology, maser technology, microwave photon detection [7], dark-matter axion search [8], quantum technologies [9,10], etc. In the context of quantum technologies, MW resonators are used to coherently drive embedded quantum systems, such as, e.g., Rydberg atoms [11], solid-state spins [12], or superconducting qubits [13]. In the specific case of spin qubits, it is the magnetic field of the MW resonator that couples to the magnetic moment associated with the spin. Here we use the enhanced magnetic field in grape dimers to efficiently drive nitrogen-vacancy (N-V) center spins in nanodiamonds (NDs). An artistic representation is shown in Fig. 1(a).

The N-V spins are solid-state quantum systems whose intrinsic coherence times can reach millisecond timescale, even at room temperature [14,15]. Their preparation and readout can be done by detecting spontaneously emitted fluorescence upon illuminating the diamond with nonresonant green light [16]. The possibility to optically probe the N-V spins at room temperature enable us to easily incorporate the quantum systems into the grape cavity. Furthermore, we consider N-V spins in diamond as the right

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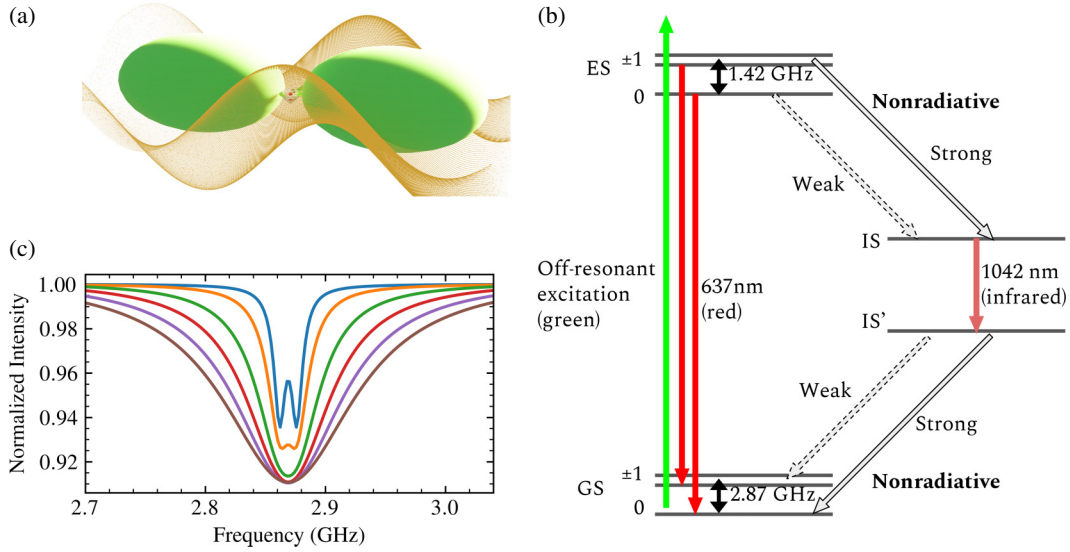


FIG. 1. (a) Artistic representation of the central idea of the present paper: a ND (red) containing an ensemble of N- V spins is placed in the gap of a grape dimer (green). The N- V spins exchange MW photons with the confined field mode between the grapes, indicated by the orange field lines. (b) N- V^- internal level structure showing the triplet (ES, GS) and intermediate singlet state (IS, IS') manifolds. The zero-field ground-state splitting (between $m_s = 0$ and $m_s = \pm 1$) is approximately 2.87 GHz. Gray-shaded arrows represent intersystem crossing (ISC) transitions ES to IS and IS' to GS. (c) Simulated ODMR profile of N- V^- for different magnetic field coupling strengths with blue representing weak coupling, orange and green representing intermediate coupling and red representing strong coupling; see the Supplemental Material [19] for calculations.

choice for this demonstration as they have proven particularly useful for sensing of magnetic fields, pressure, and temperature [17,18].

This paper is organized as follows. In Sec. II, we discuss the experimental setup for studying the coupling of N- V spins in NDs to the magnetic mode of the grape resonator and carrying out ODMR measurements to demonstrate the effect of the grape resonator. Then in Sec. III, we investigate the hotspot formation in the grape resonator using finite-element simulation. Finally, in Sec. IV, we conclude the presented study.

II. EXPERIMENTAL COUPLING OF N- V SPINS IN NDS TO GRAPE DIMER RESONATORS

We use real grapes to experimentally demonstrate improved MW field interactions with N- V spins. This section presents a brief background on N- V spin, the experimental technique used, and the measurements performed with grape dimers.

A. N- V spins

N- V spins are one of many possible defects that can exist in diamond. In its negative charge state N- V^- spins, which we denote as N- V^- spins are interesting for room-temperature quantum applications. N- V^- is a $S = 1$ system, which has triplet ground and excited states. The room-temperature energy structure of N- V^- is shown in Fig. 1(b). The ground and excited states of N- V^- are

denoted as GS and ES, respectively. Being triplet states, ground and excited states each consist of three states with magnetic spin quantum numbers $m_s = 0, \pm 1$.

Using an off-resonant optical driving with a green laser (e.g., 520 nm), N- V^- can be optically polarized to the $m_s = 0$ ground state. The green laser excites N- V^- spins from GS to ES conserving the m_s (0 to 0 and ± 1 to ± 1). The N- V^- spins relax radiatively from ES to GS by emitting a broad spectrum from approximately 600–800 nm, with a zero phonon-line transition around approximately 637 nm [20]. NV⁻ spins also undergo a nonradiative transition known as intersystem crossing (ISC) to the intermediate singlet-state manifold (labeled IS). The ISC transitions from ES ($m_s = 0$) $\rightarrow$ IS and ES ($m_s = \pm 1$) $\rightarrow$ IS occur at different rates, with the $m_s = \pm 1$ transition occurring more frequently [21]. The spin is short lived in IS compared to other transitions [22] and relaxes to IS' by emitting infrared light (approximately 1042 nm). Nonradiative transitions to the ground state IS' $\rightarrow$ GS ($m_s = 0$) and IS' $\rightarrow$ GS ($m_s = \pm 1$) also occur at different rates, with the $m_s = 0$ transition being more favorable [21]. Consequently, due to the varying rates of the ISC levels from the $m_s = 0, \pm 1$ levels, the spin eventually becomes polarized in the ground state GS ($m_s = 0$) and repeatedly cycles through excitation and de-excitation.

The states $m_s = \pm 1$ are typically degenerate in the ground state. The degeneracy can be lifted by applying a magnetic field along the N- V axis. The degeneracy can also be inherently lifted due to lattice strain [23]. The zero-field

splitting transition from $m_s = 0 \rightarrow m_s = \pm 1$ corresponds to a MW transition with frequency 2.87 GHz. With optical pumping polarizing the spin in $m_s = 0$, a MW field (near 2.87 GHz) creates a population change in the ground triplet states. Due to different rates of ISC transitions from ES ($m_s = 0$) and ES ($m_s = \pm 1$), the MW field indirectly induces a reduction in the red light emitted from the N-V spin. This process is referred to as optically detected magnetic resonance (ODMR), where magnetic dipole transitions in the MW domain are examined through the observed reduction of optical photons; see Fig. 1(c) for a typical ODMR profile with nondegenerate $m_s = \pm 1$ levels. The contrast of the ODMR plot represents the percentage reduction in N-V fluorescence intensity in the ODMR profile, depending on the strength of the MW field (see the Supplemental Material [19] for numerical modeling of ODMR and MW field strength).

B. Experimental setup

We experimentally measure the ODMR of N-V⁻ spins in NDs in the presence of grape dimers and without grapes. In order to have a small probe volume and efficient optical access, the NDs with the N-V centers are attached to the tip of a multimode optical fiber. A 1-mm diameter vertical copper wire served as an off-resonant stubbed MW antenna, driven by a MW generator (Agilent Technologies E8257D) and a 16W-amplifier (Minicircuits ZHL-16W-43-S+). The antenna was placed vertically such that the radiated component of the MW field points in the direction of the dimer axis. Here the dimer axis refers to the line passing through the major axis of each grape. As shown in Fig. 2(a), the fiber tip is positioned in between the gap between the two grapes. To a good approximation, the grapes have an ellipsoidal shape and were chosen carefully to have approximately identical dimensions, with a long axis of about 27 mm and a short axis of about 17 mm. The size of the grapes used were selected based on numerical simulations of grape dimers (see the Supplemental Material [19] for detailed materials and methods).

C. Enhanced ODMR measurements with a grape dimer resonator

ODMR measurements performed with and without grape dimers showed a clear improvement in MW coupling strength, as is evident from Fig. 2. Figure 2(b) shows two measured ODMR profiles for the case where the grapes are as close as possible to the N-V nanodiamonds (green) and the case where there are no grapes present (red). The improvement in MW coupling is inferred from the increase in contrast. The contrast was extracted from the bare ODMR measurements using a double-inverted Lorentzian fit, accounting for the strain-induced splitting between the $m_s = \pm 1$ states. Based on prior ODMR measurements on the nanodiamonds, the initial fit parameters

were set to expected values, and no boundaries or limits were imposed on the Lorentzian fit parameters during the fitting process. Lorentzian fits are known to fit N-V ODMR profiles closely [24]. Here we attribute the increase in contrast due to increases in the magnetic field strength in the gap. We note that other factors, such as laser power and temperature, could also affect the ODMR contrast. However, the laser power was stable such that the overall PL intensity counts remained stable within approximately equal to 10% throughout the experiments. As for thermal effects, since the grapes heat up due to the absorption of MW radiation, the environmental temperature exhibited by the N-V spins is expected to be greatest when grapes are present. With increasing temperature, it has been shown [25] that ODMR contrast for N-Vs in NDs decreases. Refer to the Supplemental Material [19] for more details. We, therefore, exclude temperature and laser power as potential causes for the increase in ODMR contrast, and instead attribute it to changes in the local magnetic field.

Several ODMR measurements were performed to examine the dependence of different parameters and to reveal information on the MW hotspot. With the gap size fixed to 1 mm, the grapes and fiber tip were translated at different distances from the straight copper wire antenna. The results shown in Fig. 2(c) indicate a reduction in contrast with increasing distance from the antenna. This is expected since the field strength decays away from the antenna. With the grape dimer present, an enhanced contrast was observed in all cases, indicating the presence of the grapes as the main cause for the increased contrast and not some favorable geometric arrangement. Due to additional heating when the grapes are present, the optical fiber may expand in length and move closer to the copper wire, increasing the contrast. However, this effect is negligible since each ODMR contrast measured with grapes is greater than the measured contrast without grapes within distances of 2 mm of each point. Therefore, we also eliminate thermal expansion as the cause of the increase in contrast. Furthermore, any additional fluorescence from the optical fiber itself is expected not to influence the measured ODMR contrast.

The direction of the field generated around the wire is expected to point in the same direction whether the grapes are present or not, this is supported by FEM analysis performed in Sec. III. However, the direction may differ due to experimental imperfections since FEM simulations assumed perfect ellipsoidal shapes for the dimers. The direction of the field plays a role when coupling MW fields to N-V spins. The N-V nanodiamond crystals on the fiber tip are randomly oriented, and within each crystal, there could be four possible N-V orientations. The most efficient coupling is when the MW field is orthogonal to the N-V axis [26]. Due to the randomness of the orientation of the nanodiamonds, an N-V could virtually be pointing

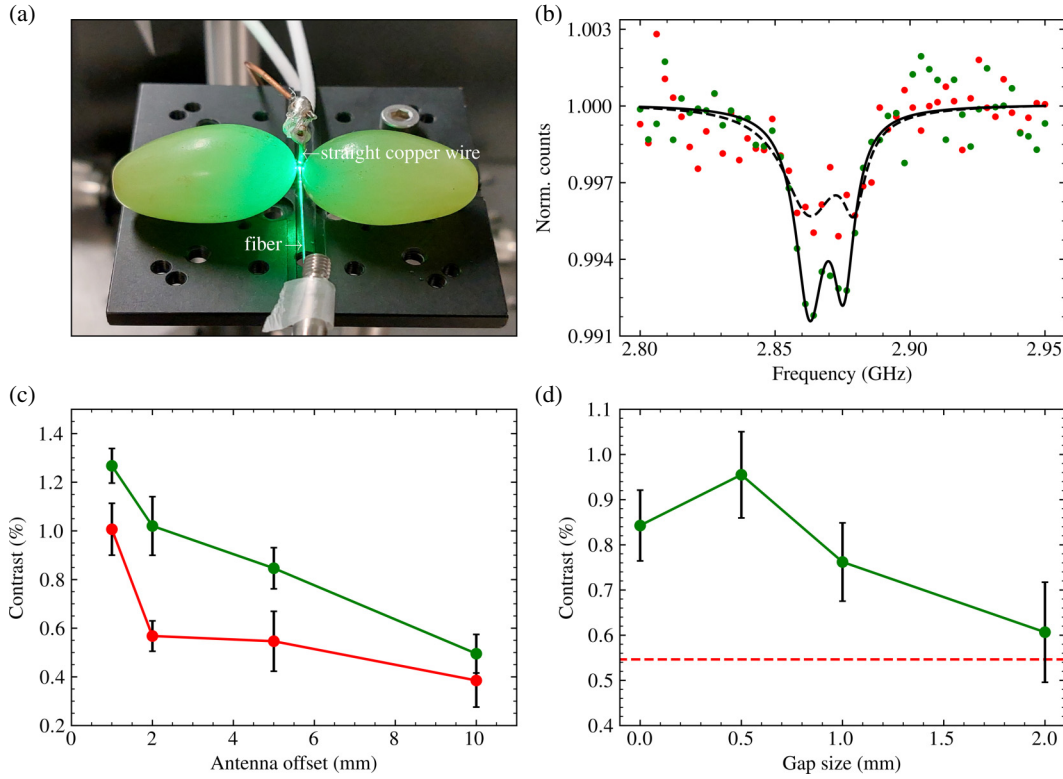


FIG. 2. (a) Photo of the experimental setup to couple MWs to N-Vs using grape dimers. A stripped optical fiber with N-V spins, cantilevered from a rod, lies between two grapes. The grapes were positioned on a platform with a vertical straight copper wire, equidistant from each grape. (b) Typical ODMR measured with (green) and without (red) grape dimers showed improved ODMR contrast. Double-Lorentzian fits (black curves) were used to extract the contrast. The dashed black curve is the fit for the case without grapes and the solid black curve is the fit for the case with grapes. (c) ODMR contrast plotted as a function of the distance from the antenna to the center of the gap, with a fixed gap size of 1 mm. (d) Contrast plotted as a function of gap size, with the distance of the wire antenna fixed to 5 mm from the gap. Error bars in the contrast were calculated using one standard deviation of the points near the flat region in the ODMR curves.

in any arbitrary three-dimensional (3D) direction. Therefore, with the nanodiamonds on the fiber tip, we measure only an average effect. We therefore attribute the enhancement in the ODMR contrast to the enhancement of the field intensity rather than the direction.

Finally, by fixing the antenna at a distance of 5 mm orthogonal to the dimer axis and varying the gap size, we were able to experimentally determine an optimum gap size of about 0.5 mm for maximum contrast improvement, see Fig. 2(d). Furthermore, we tried to explore the contrast across various points from the center of the hotspot, see the Supplemental Material [19] for more information.

For the 2-mm antenna offset case in Fig. 2(c), the largest difference in ODMR contrast was observed. In this case, the field-amplification factor that relates the ratio of the field strength with grapes to the case without grapes was estimated (refer to the Supplemental Material [19] for more details). The amplification factor for this case was estimated as $|\mathbf{B}_{\text{grapes}}|/|\mathbf{B}_{\text{no grapes}}| = 2.1 \pm 0.4$.

III. FINITE-ELEMENT SIMULATION OF MAGNETIC FIELD HOTSPOT FORMATION IN GRAPE DIMER RESONATOR

In order to verify the magnetic field hotspot between grape dimers and confirm that the increase in ODMR contrast is due to the change in the magnetic field, finite-element simulations were performed. Related work [3] investigated the MW hotspot formation in grapes and only focused on the effects of the electric field in the hotspot, which caused the plasma formation. Because the electric field is known to be large in the gap between grape dimers due to observed plasma formation, it is expected that the magnetic field should also form similar resonant behavior in the gap. Furthermore, we simulate the MW source more realistically by defining a straight copper wire antenna (as shown in the experimental setup) instead of an ideal plane wave.

Two grapes were modeled in COMSOL 6.0. Figure 3 shows the FEM results for simulated grape dimers. In the simulations, the grapes were modeled as ellipsoids and

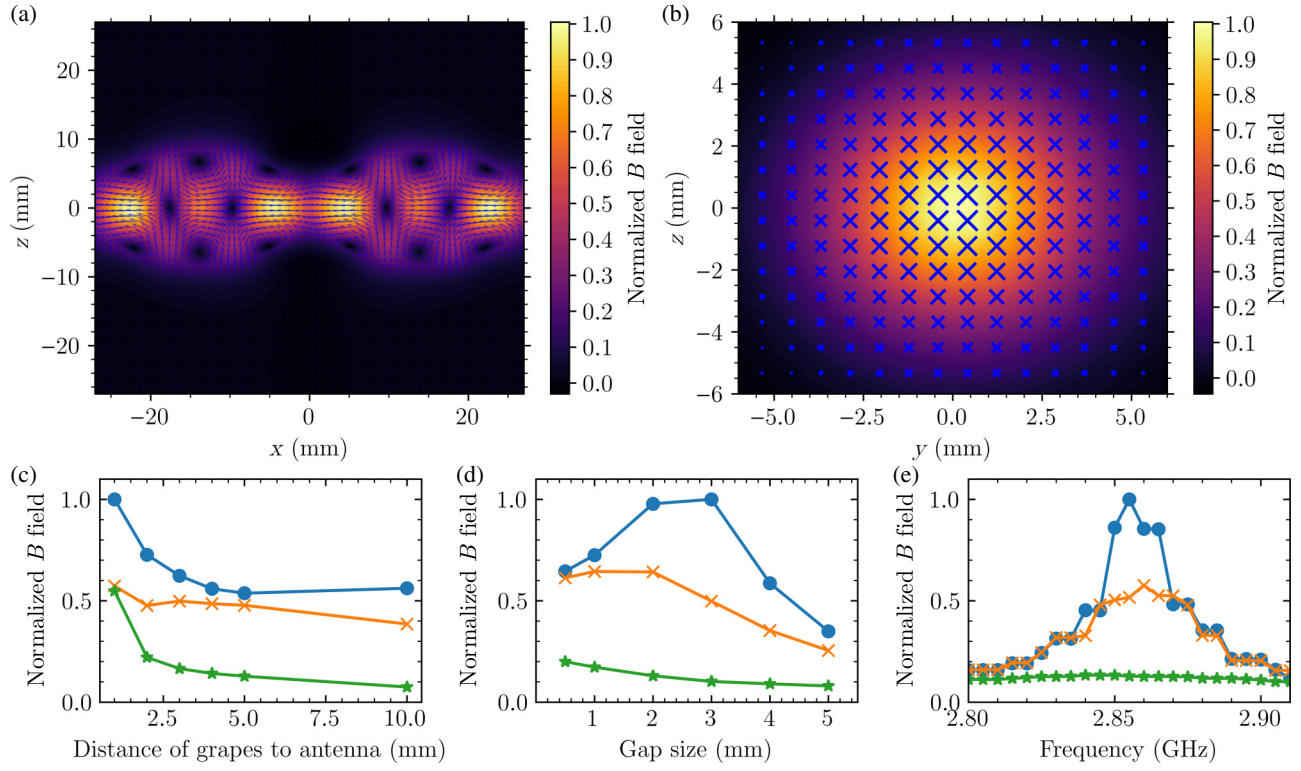


FIG. 3. Results of finite-element (FEM) simulations for an ellipsoidal dimer with dimensions of 2.7 cm (major) and 1.7 cm (minor) at 2.87 GHz. (a) The magnetic field contour and vector field at a cross section through the grapes at $z = 0$. (b) Magnetic field contour and vector field (blue crosses representing arrows into the page) at a cross section through the gap at $x = 0$ clearly shows the formation of a hotspot. (c) The magnetic field at the center of the hotspot is plotted for different antenna distances from the dimer axis. The different colors correspond to no absorption, blue ($\epsilon_r = 79.21$); weak absorption, orange ($\epsilon_r = 79.21 + 1i$); and strong absorption, green ($\epsilon_r = 79.21 + 5i$). The gap size was fixed to 1 mm. (d) The magnetic field at the center of the hotspot as a function of gap size for different absorption strengths. The antenna was fixed to a distance of 5 mm from the dimer axis. (e) The magnetic field at the center of the hotspot as a function of MW frequency for different absorption strengths. The gap size was fixed to 1 mm, and the antenna was placed at 5 mm from the dimer axis.

given the properties of water at MW frequencies ($\epsilon_r = 79.21$) as assumed by Khattak *et al.* [3]. The dimensions of the ellipsoids was taken as 27 mm (major axis length) and 17 mm (minor axis length). These dimensions were determined by measuring the major axis length of typical white seedless grapes purchased from a local supermarket. These grapes varied in size; however, a vast amount of the available grapes were approximately 27 mm long with varying widths. With approximate calculations from Mie theory, the corresponding minor axis dimensions of the ellipsoids were calculated to achieve resonance at roughly 2.87 GHz (see the Supplemental Material [19]). To deliver MW radiation to the ellipsoids, a dipole antenna with 1 mm diameter, equidistant from the dimers, was modeled to simulate a more realistic radiation source rather than just applying simple plane waves. This modeled the delivery of MW radiation more closely to the experimental setup.

The ellipsoid dimers and the antenna were embedded in an air sphere of radius 80 mm. The air sphere was surrounded by a spherical shell of thickness 53.5 mm,

which acts as a perfectly matched layer (PML). PMLs are designed to absorb radiation to prevent artificial reflections due to the finite size of the model. In this case, the PML is designed as half the maximum wavelength ($\lambda \approx 107$ mm) used in the simulation. Maxwell's equations in 3D were then solved using a frequency-domain analysis.

Figures 3(a) and 3(b) show the typical magnetic field resonance patterns at 2.87 GHz for in-plane and out-of-plane cross sections, respectively. Several maxima in the interior of the ellipsoids are observed, which is suggestive of a higher-order mode resonance. The internal field in the dimers was found to exhibit larger field strengths compared to the external field in the gap. Ideally, one can attempt to couple to these interior fields; however, the direction of the magnetic field in the interior may be difficult to control and, therefore, to couple to N-Vs. Based on Mie theory calculations (see the Supplemental Material [19]) on a dielectric sphere with a cross-sectional circumference similar to the ellipsoids, the pattern resembles a spherical

third-order magnetic field resonance mode. Since MDRs occur due to the total internal reflection of waves along the boundary, the resonance condition is typically proportional to the circular circumference of a sphere in Mie theory; a similar condition for ellipsoids is expected.

A vector-field distribution from FE simulations is also shown in Fig. 3(a), showing the internal and external vector magnetic fields. The direction of the magnetic field in the gap plays a role when coupling to solid-state spins. Angular deviations of the magnetic field will reduce the amount of coupling achieved. In the simulated ellipsoidal dimers, the magnetic field in the gap lies conveniently along the dimer axis.

Simulations were performed where the antenna was translated at various distances orthogonal to the dimer axis while remaining equidistant from each grape. In Fig. 3(c), the magnetic field at the center of the hotspot was plotted as a function of the distance to the antenna for different absorption strengths. As absorption effects increased, the magnetic field in the hotspot was weaker. The full width at half maximum (FWHM) of the hotspot was estimated from these simulations to be 6.2 mm, indicating some flexibility for placing the N-V spins for achieving enhanced coupling. As the gap size was varied, the magnetic field reached a maximum, as plotted in Fig. 3(d). The magnetic field in the gap can be optimized by tuning the gap size, in agreement with the experimental findings on gap size reported earlier. We also checked the influence of the absorption on the microwave resonances and the strength of the magnetic field hotspot. The results are displayed in Fig. 3(e). In these simulations, the gap size was fixed to 1 mm, and the dipole antenna was placed 5 mm from the dimer axis. Different absorptive levels of water were taken into account by assuming a complex electric permittivity. Since the exact absorption effects in the grapes are unknown, the complex relative electric permittivities considered were $\text{Im}(\epsilon_r) = 0i, 1i, 5i$. The peak magnetic field was found to occur at 2.855 GHz (near 2.87 GHz) with a FWHM of approximately 25 MHz (no absorption). With increasing absorption effects, more losses are expected in the media leading to a broader resonance and a smaller peak value. We also calculate the field amplification factor for the same case we estimated for the experimental measurements. The amplification factors ($|\mathbf{B}_{\text{grapes}}|/|\mathbf{B}_{\text{no grapes}}|$) for three different absorptions simulated were found to be 6.2, 4.1, and 1.9 for $\text{Im}(\epsilon_r) = 0i, 1i, 5i$, respectively. Given the imperfections in the real grapes and the presence of absorption, the measured amplification factor of 2.1 lies within the calculated amplification factor from simulations.

Comparison between the simulation results and the experimental findings gives a consistent picture: the formation of a magnetic field hotspot between the grapes in our FE simulations nicely explains the measured increases

in ODMR contrast as a function of gap size and spatial location.

IV. CONCLUSIONS

We studied microwave hotspot formation in grape dimers using ODMR measurements of N-V spins in diamond at room temperature. We observed enhanced ODMR contrasts with the grape dimers compared to the case without them. Using FEM simulations we attributed the observed enhancement of ODMR contrasts to the magnetic field hotspots within the gap of the grape dimers. Our findings extend the work by Khattak *et al.* [3], which explored plasma ignition in grape dimers within microwave ovens.

Microwave resonators play a role in many different applications and a key example is the room-temperature masers using solid-state spin systems [27–32]. The study presented in this paper lays the foundation for exploring room-temperature microwave cavity designs based on dielectric multispherical and ellipsoidal geometries for such maser platforms or in general spin-based quantum technologies. The microwave resonance in the case of grape dimers is resulting from the water content in the grapes. Specifically, water has a high refractive index (approximately equal to 9) in the microwave domain, compared to alternative materials like sapphire (approximately equal to 3), which is typically used in MW resonators. This means for some resonator design frequency, a water-based cavity design will be much smaller in size. However, a challenge for maser applications is the Q factor, which is crucial for sustaining microwave stimulated emission. In the presented grape dimer case, simulations indicate a linewidth of 25 MHz at 2.855 GHz, corresponding to a Q factor of approximately 114. This is significantly lower than the Q factors typically required for maser resonators, which are generally in the range of 10^4 to 10^6 [33]. In practice, the dielectric losses in water contribute to this lower Q factor, as water is inherently more lossy than materials like sapphire [34,35]. Additionally, we are coupling through the evanescent field between the grapes, which may limit the Q factor. Future work could explore incorporating composite materials with high-permittivity, low-loss dielectrics into the water. These materials could potentially enhance the performance of water-based resonators by improving both the Q factor and reducing energy dissipation, leading to more efficient designs for maser applications. By optimizing resonator geometries and refining material properties, these water-based designs could offer a trade-off between compactness and efficiency, opening up possibilities for more compact and efficient on-chip maser platforms.

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T.V. conceived the central idea of the project; T.V. and S.R. designed the project. S.R. and A.F. designed and built the experimental setup; A.F. and S.R. performed the experiments; A.F. and S.R. analyzed data; A.F. and S.R. performed theoretical modelling; A.F. conducted finite-element modeling. All authors interpreted the results and wrote the manuscript. S.R. and T.V. supervised the project.

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Correction: The institutional name in the second affiliation in the front matter has been corrected. The first sentence of the Acknowledgments and the seventh sentence of the second paragraph of Sec. IV contained typographical errors and have been corrected.