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ABSTRACT
We demonstrate the cooling of a microwave mode at 2872 MHz through its interaction with optically spin-polarized NV\textsuperscript{−} centers in diamond at zero applied magnetic field, removing thermal photons from the mode. By photo-exciting (pumping) a brilliant-cut red diamond jewel with a continuous-wave 532-nm laser, outputting 2 W, the microwave mode is cooled down to a noise temperature of 188 K. This noise temperature can be preserved continuously for as long as the diamond is optically excited and kept cool. The latter requirement restricted operation out to 10 ms in our preliminary setup. The mode-cooling performance of NV\textsuperscript{−} diamond is directly compared against that of pentacene-doped \textit{para}-terphenyl, where we find that the former affords the advantages of cooling immediately upon light excitation (whereas pentacene-doped \textit{para}-terphenyl undesirably mases before it begins cooling) and being able to cool continuously at substantially lower optical pump power.

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A century ago, the invention of superheterodyning substantially enhanced the sensitivity with which weak radio signals could be detected in the face of noise. Today, after many decades of research into semiconducting materials and devices, low-noise amplifiers operating at GHz frequencies with large fractional bandwidths can be readily purchased. These amplifiers, typically in the form of HEMT-based MMICs, require only a source of low-voltage dc power to operate and offer noise figures as low as ~0.3 dB, corresponding to amplifier noise temperatures of just a few tens of kelvin. This fantastic technological achievement has left us in the situation where the sensitivity at which rf measurements can be made at room temperature is almost entirely limited by thermal (i.e., Johnson–Nyquist) noise. The obvious route to further improving sensitivity and attaining the single-photon (also known as “quantum”) limit is to cool the entire instrument/experiment down within a refrigerator. For measurements at microwave frequencies, this means cooling to mK temperatures by means of a dilution refrigerator. However, the machinery is bulky, fragile, and power hungry (consuming kilowatts, overall) while offering minuscule cooling powers (milliwatts, at best). Alas, nobody has yet managed to miniaturize a dilution refrigerator to fit and operate (off batteries) within the form factor of a mobile phone or wrist watch.

A recent paper explored a radical alternative to physical refrigeration of the instrument. Instead, a single microwave mode is cooled, reducing the number of thermal microwave photons occupying it and, thus, reducing the amount of Johnson noise, in units of watts per Hz of bandwidth, extracted from it. This more targeted form of cooling was achieved through the mode’s interaction with a strongly spin-polarized material exhibiting a cryogenic spin temperature across a paramagnetic transition in tune with the mode. The material in question was an organic molecular crystal, namely, pentacene-doped \textit{para}-terphenyl (Pc:PTP), whose spin-coldness was generated by optical pumping. This demonstrator suffered from initially masing after the application of a high-power (and high overall energy) optical pulse before its mode-cooling effects could be accessed, as well as only being able to operate in pulsed mode, cooling the microwave mode for just a few hundreds of microseconds at a time. In this Letter, we report on the sustained cooling of a microwave mode, achieved using photo-excited negatively charged nitrogen vacancies (NV\textsuperscript{−}) in diamond as the cryo-spin-polarized absorber instead. The setup is run entirely at
zero dc applied magnetic field (ZF) and under ambient temperature on the lab benchtop. Because the setup operates at ZF, there is no need to set up strong magnets or align the diamond crystal to a magnetic field (no orientation dependence). NV– diamond differs from Pc:PTP in many ways; most pertinently, it requires substantially lower optical pumping power to become spin-polarized to an equivalent extent. In this study, we describe the mechanism, anatomy, and performance of our mode cooler and compare the relative merits of NV– diamond to Pc:PTP in this application.

Our experiment uses a round brilliant cut diamond with a girdle diameter of 2.65 mm, a table size of 2 mm, a total height of 1.5 mm, and a pavilion angle of 41.5°. Though natural (i.e., mined from the ground), the diamond had been “color enhanced” through annealing. It has a dark red color and fluoresces bright red under 532 nm illumination. From UV/Vis spectroscopy shown in the supplementary material, the sample absorbed broadly at 576 nm and sharply at 638 nm, the latter being the characteristic zero-phonon line of NV– centers.

At ZF, the frequency corresponding to the energy difference between the lowest energy level (|0⟩, corresponding to m_s = 0) and the upper pair of degenerate levels (|1⟩, |2⟩, corresponding to m_s = ±1) is 2870 MHz. The spin-dependency of the ISC rates acts to increase the number of NV– centers in the |0⟩ level relative to the |1⟩ and |2⟩ levels, so causing the transitions between |1⟩ or |2⟩ and |0⟩ to become absorptively hyperpolarized. Additionally, the central microwave resonance at 2870 MHz for a bulk sample of NV– centers in diamond will be split into two distinct resonance peaks due to the effects of local electric fields. Measurements of our sample using a homemade photoexcited transient electron paramagnetic resonance spectrometer at ZF (ZF-trEPR) confirm this splitting of the resonance line with the strongest observed ZF-trEPR signals in the vicinities of 2867 and 2873 MHz, see Fig. 2. Similar resonance splittings have been observed in ZF ODMR of NV– diamond.

Mode cooling can be performed by tuning a sufficiently high-Q microwave mode to the frequency of either of the two absorptive peaks. We measured the mode-cooling response at the same discrete nine interrogating frequencies as used for ZF-trEPR and found that the cooling was deepest when the microwave mode was tuned to 2872 MHz. The mode cooling at 2867 MHz was not quite as deep as that for 2872 MHz, but far deeper than that at 2870 MHz. We thereupon chose f_{mode} = 2872 MHz as the frequency at which to study the cooling in detail; this is indicated as the transition frequency of |0⟩ → |2⟩ in Fig. 1(a).

In a similar manner to how the X → Z triplet transition of Pc:PTP in our previous work was used to cool a microwave mode at 1450 MHz, we here exploit the |0⟩ → |2⟩ transition in NV– diamond for mode cooling. We use a similar setup as in the previous work, incorporating a high-gain superheterodyne receiver to measure the

![Diagram](image-url)
SAW bandpass filter used before the detector in our previous work\(^2\) is to a wider –3 dB linewidth of 1 MHz, the original 50-kHz-bandwidth eight in volume) with a lower loaded quality factor (\(Q_L\)) of 2900. A STO dielectric resonator is now almost half of its previous size (one \(8\)th in the copper cavity with the beam focused down to a spot diameter of \(1.5 \text{ mm at the sample.}\)

Finally, since the lower loaded \(Q_L\) corresponds to a wider –3 dB linewidth of 1 MHz, the original 50-kHz-bandwidth SAW bandpass filter used before the detector in our previous work\(^1\) is replaced with a 1.25-MHz-bandwidth version; this avoids excessive temporal delay and distortion of the detected power signal (albeit at the expense of allowing a small fraction of noise on the shoulder’s of the resonator’s line profile into the superheterodyne signal path). As seen in Fig. 2(b), the resonator’s linewidth is narrow enough to ensure that only the NV\(^{-}\) center resonant at 2872 MHz, to which the resonator is coupled (indicated by a red marker), and the shaded region shows the SAW filter bandwidth.

Instantaneous microwave power extracted from the microwave mode in a cavity, monitored on an oscilloscope (LeCroy DDA-260, DC-coupled) triggered by the optical pumping pulse.\(^3\) A few modifications are worth noting: to be resonant at the higher frequency of 2872 MHz, the STO dielectric resonator is now almost half of its previous size (one \(8\)th in volume) with a lower loaded quality factor (\(Q_L\)) of 2900. A microwave iris (stub-tuner) has been added between the resonator and LNA to allow precise matching of the cavity to critical coupling (reflection coefficient \(S_21 = 0\)). The optical pump source is a 2-W continuous-wave 532-nm diode-pumped Nd:YAG solid-state laser, whose output is gated using square pulses from a pulse generator. The NV\(^{-}\) diamond, placed inside the STO resonator within a copper cavity as depicted in Fig. 1(b), is excited by the laser from above through a hole in the copper cavity with the beam focused down to a spot diameter of \(1.5 \text{ mm at the sample.}\)

The cooling signal then decays back to the baseline noise power (indicated by \(\Delta P = 0\)) with a decay time of about 10 ms after the laser pulse ends. The temperature decrease and time dynamics of the signal were simulated in Fig. 3(b), where \(T_{\text{mode}}\) is simulated from solving coupled differential equations dictating NV\(^{-}\) diamond’s spin dynamics and \(q\) (see the supplementary material).\(^4\)\(^–\)\(^1\)\(^5\)

We here compare the mode-cooling capabilities of NV\(^{-}\) diamond to those of Pc:PTP as directly as possible. At ZF, both systems cool the microwave mode through stimulated absorption, but NV\(^{-}\) diamond has an obvious advantage in that it immediately cools the microwave mode after pumping. Pc:PTP undesirably mases first (through stimulated emission) after pumping, during which it cannot cool the mode (see the supplementary material). Only after the masing period ends can Pc:PTP cool the mode through stimulated absorption.\(^2\) Furthermore, the \(0 \rightarrow |2\rangle\) transition for NV\(^{-}\) diamond exhibits no crossover (from absorptive to emissive) at later times; it can only cool at ZF. In Fig. 3(d), the cooling signal from our present experiment is compared against an adaptation of a measured cooling signal for Pc:PTP (post-masing) reported in our previous work,\(^6\) where Pc:PTP is shown to be able to cool the mode to a much lower temperature (50 K), but requires high-power pulsed excitation lasting only 300 \(\mu\text{s}\) at \(~5-KW\) peak pulse power. Due to the pulsed nature of the excitation, the cooling response of Pc:PTP does not last more than 0.5 ms. Conversely, the cooling afforded by NV\(^{-}\) diamond, even from a pulsed excitation, lasts up to 10 ms, which is longer than that attained with Pc:PTP by two orders of magnitude. Furthermore, compared to the dye laser used for pumping Pc:PTP, the 2-W Nd:YAG laser used here is substantially cheaper and easier to purchase and maintain. The disadvantage of NV\(^{-}\) diamond though is its inferior depth of the cooling. Methods to overcome this will be discussed toward the end of this Letter.

Attempts at pumping Pc:PTP with the same 2-ms 532-nm laser pulse used for NV\(^{-}\) diamond (after switching back to an STO resonator tuned to 1450 MHz) produced no cooling response. Likewise, we attempted to excite NV\(^{-}\) diamond using Q-switched 5.5 ns high power pulses from an optical parametric oscillator (Litron Aurora II Integra) at 532 nm. This gave a much smaller power reduction of only 0.4 dB, which corresponds to much weaker cooling with \(T_{\text{mode}}\) dropping by \(30 \text{ K only.}\) We note that NV\(^{-}\) diamond produces a lower polarization if pumped with pulse lengths shorter than the decay times of \(|S\rangle\) to \(3A_2\) (\(\sim 1 \mu\text{s}\)),\(^2\) so this may explain why the nanosecond pump pulses produce a poor cooling result. Overall, the mode cooling afforded by

\[
q = (\exp(\frac{hf_{\text{mode}}}{k_B T_{\text{mode}}}) - 1)^{-1}.
\]

In Eq. (1), \(f_{\text{mode}}\) is the mode frequency at 2872 MHz. Cooling the mode down to 192 K represents a reduction from 2103 to 1392 in the number of thermal photons.

The cooling signal when decayed back to the baseline noise power (indicated by \(\Delta P = 0\)) with a decay time of about 10 ms after the laser pulse ends. The temperature decrease and time dynamics of the signal were simulated in Fig. 3(b), where \(T_{\text{mode}}\) is simulated from solving coupled differential equations dictating NV\(^{-}\) diamond’s spin dynamics and \(q\) (see the supplementary material).\(^4\)\(^–\)\(^1\)\(^5\)

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\[
q = (\exp(\frac{hf_{\text{mode}}}{k_B T_{\text{mode}}}) - 1)^{-1}.
\]
Pc:PTP is deep but short, whereas that afforded by NV\textsuperscript{-}/C\textsubscript{0} diamond is not as deep but long, while requiring vastly lower instantaneous optical pump power.

After having demonstrated how NV\textsuperscript{-}/C\textsubscript{0} diamond could cool the microwave mode under pulsed excitation, we investigated whether the microwave mode could be cooled continuously for as long as the NV\textsuperscript{-}/C\textsubscript{0} diamond was optically pumped, beyond its relaxation time [about 10 ms in Fig. 3(a)]. Figure 4(a) shows the noise power reduction when the NV\textsuperscript{-}/C\textsubscript{0} diamond is pumped by a 10-ms square laser pulse (with a repetition frequency of 6 Hz). The resultant signal can be seen to follow the time profile of the laser excitation, where the mode is maintained at its cooled temperature for as long as the laser pulse is on. From Fig. 4(b), the experimental $T_{\text{mode}}$ mode is calculated to be cooled down to and held at the temperature of 188 K (reducing the number of thermal photons to 1363). After the laser pulse, the system relaxes with the same decay time as in Fig. 3(a). We were similarly able to simulate the dynamics of $T_{\text{mode}}$ in Fig. 4(b) with identical parameters to those used for simulating Fig. 3(b) but with a longer laser pulse. This then demonstrates the capability of the material to continuously cool a microwave mode at ZF so long as it is spin-polarized (through optical excitation). Being able to continuously cool a microwave mode is a significant advantage that NV\textsuperscript{-}/C\textsubscript{0} diamond has over Pc:PTP, which can only cool in a pulsed manner.

Though we have demonstrated how NV\textsuperscript{-}/C\textsubscript{0} diamond can be used to continuously cool a microwave mode, improvements are needed for the effect to be more usefully exploited. Using a bigger diamond shaped as a cylindrical rod (4-mm diameter) could improve upon the resonator’s magnetic filling factor. Combined with increasing the concentration of NV\textsuperscript{-} centers and boosting the excitation intensity of the pump laser, we judge that the number of spin-polarized NV\textsuperscript{-} centers
interacting with the TE$_{013}$ could be straightforwardly increased by at least an order of magnitude. A rod shaped sample (with polished flat end windows) would prevent excessive retro-reflection of pumping light compared to the brilliant cut of our current sample, which is expressly designed to reflect light for visual appeal. Tradeoffs and diminishing returns admittedly lurk here: (i) higher concentrations of NV$^-$ necessitate higher concentrations of substitutional nitrogen defects, which have dipolar coupling interactions with NV$^-$ centers leading to line broadening and shorter $T_2^*$; (ii) higher optical pump power would increase polarization, yet carry the risk of converting NV$^-$ centers into NV$^0$ centers, so reducing the number of the former available for mode cooling. We note that, empirically, the insertion of an ND filter across the laser’s pump beam (so as to attenuate it) would always reduce the observed depth of mode cooling, suggesting that our current setup would have immediately benefited from greater pump power (up to a point).

The cooling of the microwave mode could be made truly continuous by actively cooling the diamond and resonator through one or a combination of: (i) solid thermal anchorage (heat-sinking), (ii) forced air, (iii) immersive liquid cooling (locating the diamond within a “flow tube”), or (iv) heat pipes, which are all still less costly to implement and maintain than dilution refrigeration. Given the high thermal diffusivity of diamond and the relatively modest absorbed optical pump power in need of being removed, the engineering challenge of constructing a suitable cooling system appears highly feasible.

In conclusion, we have demonstrated how NV$^-$ diamond at ZF can be used to cool a microwave mode down to 188 K and hold it at that temperature continuously for as long as the diamond is optically excited and kept cool. The mode cooling performance of NV$^-$ diamond was then compared to that of Pc:PTP, where NV$^-$ diamond demonstrates the advantages of cooling continuously under excitation, requiring much lower excitation power, and cooling the mode immediately upon excitation to boot (in contrast to Pc:PTP, which mases before cooling). Though, in our current implementation, NV$^-$ diamond could not cool the mode to as low a temperature as Pc:PTP, we propose multiple avenues by which the cooling performance of NV$^-$ diamond could be improved.

See the supplementary material for additional experimental details, the UV/Vis spectrum of NV$^-$ diamond, further comparison between NV$^-$ diamond and Pc:PTP signals, NV$^-$ diamond spin dynamics simulation, and noise analysis.

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**AUTHOR DECLARATIONS**

**Conflict of Interest**

The authors have no conflicts to disclose.

**DATA AVAILABILITY**

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

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