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Nanoscale thermal imaging with spins in diamond - ThermoQuant

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ABSTRACT

Temperature sensing from the macroscopic world to the nanoscale find widespread applications from the life-sciences to electronic circuit design and beyond. Solid-state electronic spins based on nitrogen-vacancy centers in diamond have the potential to be temperature sensors with sensitivities in the K/\sqrt{Hz} -range and spatial resolutions down to 10 nm. However, the high thermal conductivity of diamond constitutes an impediment for such non-invasive temperature measurements. Throughout the project, experimental and numerical analyses were investigated with different geometrical configurations including bulk diamond, quartz, heterostructures of both materials and nano-diamonds dispersed on quartz, suggesting that engineered heterostructures provide a valuable option for non-invasive thermal imaging.

Keywords: diamond; NV center; thermal imaging, thermal sensing.

1. INTRODUCTION

Thermal sensing and imaging at the nanoscale, has become a key aspect in a broad range of fields like materials science, electronics, chemistry, and life sciences. Several approaches are being explored including tip-enhanced infrared or Raman thermometry [1, 2], scanning thermal microscopy (SThM) [3, 4], SQUID-based nano-thermometry [5], and nanoscale fluorescence thermometry [6]. However, none of these techniques offer fast, sensitive (K/\sqrt{Hz} range), and quantitative imaging with sub-micron spatial resolution under ambient conditions. Using Nitrogen-Vacancy (NV) electronic spins in diamond as thermal sensors, in a scanning probe configuration not only provides all these characteristics but is also of high importance in this field because:

- It can improve the sensitivity and resolution of the current state-of-the-art solutions by a factor of 10.
- It can respond to the pressing need in the semiconductor and data storage industries in going beyond Moore's law.
- It can represent a key usage of quantum technology to overcome measurement barriers that are physically imposed by classical technologies.

The use of electronic spins in diamond as thermal sensors is a breakthrough innovation in nanoscale thermal imaging. It brings significant potential for advances in science and technology, where it can stimulate development in fundamental physics, material research and in the life-sciences. Specifically, in materials science and mesoscopic physics, it may open new research avenues by enabling direct access to heat production on the nanoscale.

The collaboration between the French CNRS Laboratory Charles Coulomb (CNRS) and the Swiss quantum sensing start-up company Qnami (QNAMI) within this project led to the following results:

- An experimental proof-of-concept for thermal imaging using quadriwave lateral shearing interferometry [7].
- An advanced design for a quantum sensor for thermometry, together with simulations.
- A patent search and filing.
- A scientific publication [8].
- The production of a first, scanning-probe-based, prototype.

2. STATE OF THE ART

There is currently only one available roomtemperature technology for measuring temperature at the nanoscale: SThM. This technique is based on Atomic Force Microscopy (AFM) and reads thermal signals by using specialized probes, which contain a resistive element that is brought in contact with the sample of interest. Temperature variations are obtained through a measurement of the change in resistance of the sensor. This technique has been widely used in different fields of application such as failure analysis of microelectronic components, where it offers sub-100 nm resolution, but suffers from several important drawbacks such as the need for frequent replacement of the consumable element, the impossibility to perform quantitative measurements, a small dynamic range and the impossibility to measure temperature in materials with high thermal conductivity [3, 4].

There are other solutions for ambient nanoscale thermometry which are being developed but are not commercially available. An example particularly worth mentioning are nano-diamonds that can be directly deposited onto samples of interest for thermal sensing with NV electron spins [9]. This approach is quantitative and delivers one of the highest thermal sensitivities reported so far under ambient conditions. However, a key drawback is the random positioning of the nanodiamonds and the resulting poor spatial resolution and reproducibility.

The NV based thermometry technique explored within the ThermoQuant project promises to improve drastically on sensitivity and resolution. Moreover, the nano-probes are highly robust and readily compatible with the commercial quantum sensing platform "Qnami ProteusQ" which was launched by Qnami in 2019.

3. BREAKTHROUGH CHARACTER OF THE PROJECT

NV centers in diamond are extremely useful for sensing and imaging due to the possibility to detect their electron spin resonance optically [10]. This characteristic enabled the creation of highly sensitive NV-based quantum sensors, that can probe properties such as: strain [11, 12], electric [13, 14] and magnetic fields [15-17]. Additionally, the electron spin resonance of the NV center has been used for thermometry relying on the temperature-dependant shifts of the NV's energy levels [18]. The use of this temperature shift on the electron spin sublevels has been demonstrated using single NV centers embedded in nano-diamonds and can reach a sensitivity of 100 mK/\sqrt{Hz} [9]. There are several ways in which this thermometry technique can be used. The first one consists in grafting an NV-doped nano-diamond at the apex of an optical fiber [19] or an AFM tip [20, 21], which is later used to scan over the surface of interest. The main drawback of this technique is the complexity of the fabrication process, together with the short spin coherence times that are typical of NV centers in nano-diamonds. Secondly, these nano-diamonds can

be dispersed on the surface of interest and can be individually imaged using a wide-field detection scheme [9, 22, 23]. However, the random position of the thermal sensors on the surface limits the quality of the probe and therefore does not respond to the needs of applications in, for example, the data-storage or the semiconductor industry.

These issues can be addressed by using a radically new approach in the fabrication of nanoscale thermal probes, using techniques and processes from the microelectronics industry. This approach maximises robustness, sensitivity, and resolution, as it has been previously demonstrated for nanoscale magnetic imaging [24]. Nevertheless, fundamental challenges arise with this approach due to diamond's high thermal conductivity. One of the main goals of this project was to overcome these challenges, and thereby set a solid starting point for the future implementation and commercialisation of this highly promising, novel thermal imaging technology.

4. PROJECT RESULTS

Thermal imaging using a wide-field scheme was performed on nanoplasmonic structures fabricated by QNAMI on diamond and quartz. These experimental results were then compared to numerical simulations and used as a basis for the experimental assessment of more complex, nanostructured diamond thermometers. The experiments were performed by CNRS using lateral quadriwave shearing interferometry [7].

The nanoplasmonic structures, which are used as heat sources in this experimentation, consist of an array of gold nanoparticles illuminated at their plasmonic resonance wavelength [25, 26].



Fig. 1. Maximum local heating δT_{max} as a function of the absorbed optical power P_{abs} for thermoplasmonic arrays deposited on quartz (blue squares) and diamonds (red dots); insets: temperature maps recorded (i) on the quartz substrate for $P_{abs} = 2.9$ mW leading to $\delta T_{max} = 115.2$ °C (top within the blue frame) and (ii) on the diamond substrate for an absorbed laser power of $P_{abs} = 68.8$ mW leading to $\delta T_{max} = 12.8$ °C (bottom within the red frame); solid lines are linear fits of the experimental data.

Further details of these structures, their operation, the experimental conditions, and the basic principle of how the thermoplasmonic structures work as heating structures can be found on the scientific publication that was one of our key achievements within project ThermoQuant [8]. The main results for the thermal imaging of these heating structures can be seen in Fig. 1. The data shows that the maximum local heating δT_{max} is linearly proportional to the absorbed power P_{abs} for both substrates used, as expected. Nevertheless, the heating induced by the nanoplasmonic array on the diamond substrate is much lower than the one observed on the quartz substrate. A difference of 2 orders of magnitude can be observed, with a maximum heating per absorbed power of 0.2 K/mW and 38 K/mW for diamond and quartz substrates, respectively. This difference in heating is caused by the difference in thermal conductivity of the substrates, which is about 3 orders of magnitude higher in diamond than in quartz. Therefore, heat transfer in diamond is very efficient, while it is much reduced in quartz. Consequently, the diamond substrate acts as a thermal sink that cools the heat source [27]. As such, any thermal imaging structure based on bulk diamond will be highly invasive as the measured temperature of the heat source will be reduced by the diamond substrate.

A possible mitigation strategy to reduce the heat dissipation in bulk diamond would be to create a substrate composed of a small volume of diamond and a low thermal conductivity substrate, such as quartz.



Fig. 2. Numerical simulations of the maximum local heating δT_{max} at the sample surface induced by a two-dimensional $10 \times 10 \ \mu m^2$ heater for increasing heat powers P_{heat} ; four configurations are considered: the hot plate located either on a diamond substrate (red dash line), on a quartz substrate (blue solid line), on a 100-nm thick diamond membrane bonded on a quartz substrate (orange dotted line), or on a quartz substrate with diamond nanostructures dispersed on the surface (green dots); in this latter architecture, temperature increase is the one reached within the nanostructures whose dimensions are in the range of few tens of nanometers; the insets show schematics of the four configurations; the temperature increase per heating power reaches ≈4 K/mW in the heterostructure geometry; it is almost two orders of magnitude higher than the one predicted on bulk diamonds and is solely ≈ 6 or 7 times smaller than the one expected on the quartz geometry.

Fig. 2 shows finite-difference time-domain (FDTD) simulations based on a heat conduction modelling of temperature increase as a function of heat power for three different diamond-based architectures: (i) a bulk diamond, (ii) a heterostructure composed of a 100-nm thin diamond membrane on quartz, and (iii) nanodiamonds dispersed on the heat source. The achieved temperature increase in the diamond-quartz heterostructure is much higher than the one for a bulk diamond but is still lower than that of quartz. This arises from the reduced effective thermal conductivity of the heterostructure. On the other hand, the temperature of nano-diamonds exactly matches the one expected on the thermal conductivity quartz substrate. This low thermalization of the nano-diamonds allows for a nonperturbative measurement of the heat source temperature, even though the nano-diamonds can be randomly positioned. These results show that engineered heterostructures, such as stacking thin diamond membranes over low conductive substrates, provide significantly reduced heat diffusion, and appear to be a valuable option for micron-scale thermal imaging using NV spins in diamond.

Based on the idea of these heterostructures, QNAMI proposed designs for scanning-probe-based sensors using the same principle, which were then simulated to find an optimal solution, focused on geometry, doping and isotopical engineering of the diamond structure. Finally, the optimal heterostructure was chosen to produce the first prototype, which included the compatibility with the Qnami ProteusQ.

5. FUTURE PROJECT VISION

5.1. Technology Scaling

The Qnami ProteusQ is a complete quantum microscope system and is the first commercial scanning NV microscope. Having realised first prototypes of our novel nanoscale thermal sensors with a compatibility with this readily available product, will enable the validation of the technology in a relevant customer environment.

5.2. Project Synergies and Outreach

As mentioned before, the compatibility of the developed sensor with the Qnami ProteusQ is one of the key components for further validation and scaling of the project outcome. Once the patent for the project is accepted, it will be public and will also facilitate the dissemination of our ATTRACT results and activities.

5.3. Technology application and demonstration cases

The first demonstration case is going to be on the data storage and semiconductor industries. An emerging trend in further scaling in data storage is heat-assisted magnetic recording (HAMR), where nanometer-scale hotspots are used to enable fast and reliable switching of nanoscale bits. Direct visualisation of thermal profiles in these devices is a key bottleneck for R&D and a timely industry-application this technique will address. The semiconductor industry, on the other hand, would be a second demonstration as it is in urgent need for strategies to go beyond Moore's law. This means replacing brute force scaling down of silicon technology with hybrid designs that optimize lifetime, power consumption and functionalities. Yet dimensions remain small and in practice these developments demand fine design analysis at a 50 nm scale and below. Temperature management has been identified as one of the key challenges for this development and other hight-tech applications.

The nanoscale thermal imaging and sensing technologies developed within ThermoQuant will advance fundamental science and transfer NV-based quantum sensing from the laboratory to advanced applications. ThermoQuant will thereby yield several societal and economic benefits such as:

- An opportunity for European semiconductor and data storage industries to gain shares in a market that is currently dominated by US and Asian companies.
- Smaller, more energy efficient electronic devices for European citizens, which our thermal imaging technology will ultimately enable.
- Optimized batteries and battery management.
- A novel medical diagnostic tool, potentially applicable, e.g. to early stage cancer diagnosis.
- A further strengthening of European quantum industry, including new job opportunities.
- A strengthened, European quantum ecosystem, which may have a strong, transformative effect on society in the long term.

5.4. Technology commercialization

The initial steps taken for the commercialization of the technology were taken by Qnami in 2019 when the Qnami ProteusQ was launched. This scanning NV microscope is used for the analysis of magnetic materials at the atomic scale but the electron spin in NV centers can be used to analyse temperature as well. Therefore, by creating a thermal sensor that is compatible with this readily available product, the commercialization and distribution will be much quicker.

5.5. Envisioned risks

As a new technology increases its readiness level, the need for testing in industrially relevant environments makes performance requirements stricter, especially in a rapid changing and improving industry such as semiconductors and data storage. Therefore, continuous improvement of the product would probably be required, which can be time consuming. Nevertheless, by the time a possible ATTRACT Phase 2 project is started, several Qnami ProteusQ users would be available and testing this new technology with many of them will represent a speed-up in feedback which in turn will provide faster further improvement of the technology.

5.6. Liaison with Student Teams and Socio-Economic Study

Collaborating with MSc. Level student teams could be very beneficial for the project, as they would provide ideas in which the project could address societal challenges. Prof. Patrick Maletinsky, co-founder of QNAMI and Prof. Vincent Jacques from CNRS have previous experience with numerous MSc. Level students within their research groups. Additionally, QNAMI has had experience with MSc. And BSc. Level students and master thesis projects (MSc. Marcelo Gonzalez, BSc. Agathe Juneau), where one of those students finished his thesis and obtained an engineering job at the company. Having this type of people on the consortium would facilitate the explanation material of the technology. The contribution from these students to the project during a possible ATTRACT Phase 2 would provide a broad perspective of how the technology can help solve different challenges and the consortium would be willing to provide these perspectives to the expert-driven socioeconomic study through interviews or publications that are made during the project.

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