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Doctoral thesis review of reviewer Ronald Ulbricht

Title of the doctoral thesis:
Magnetic field imaging in biological systems with nanometric resolution
by
Ing. Josef Souček

1. Topicality of the dissertation topic

The thesis sets out to develop an on-chip imaging platform for measuring magnetic signals in biological systems using quantum sensing methods that employ nitrogen-vacancy centers in diamond. Both photoluminescence (PL) and photocurrent readout methods are to be employed. A successful implementation could be useful for biomedical applications.

2. Fulfilment of the objectives of the dissertation thesis

Two main objectives were defined: 1) modelling and experimental verification of charge carrier dynamics of NV centers and their interaction with the diamond host, and 2) design, fabrication and application of on-chip magnetic field sensing. All in all, one can say that the objectives were met. I find the title part 'with nanometric resolution' however somewhat overselling of what is actually presented. I can understand that this is probably a long-term goal, but with a title like that I would have expected it to be demonstrated already. It is not even mentioned how 'nanometric resolution' is supposed to be achieved in theory or how it is actually defined.

3. The methods and procedures of the solution

Photoelectric detection of magnetic resonance (PDMR) of NV centers is a rather new method to perform quantum sensing with NV centers and was pioneered by the Hasselt group. PDMR involves more processes compared to the more standard optical counterpart ODMR, such as deliberate ionization of NV that creates charge carriers in the diamond electronic bands. To gain more understanding, in this thesis work numerical simulations based on sets of rate equations were solved in space and time to model the dynamics of charge carriers in the diamond bands as created by photoionization of defects and their recombination, which in turn leads to charge state conversion of defects. The interplay of these effects together with the internal electronic population dynamics of the defects ultimately determine key parameters such as NV- spin contrast and the simulations attempt to capture this. Some of the theoretical results were compared to experimental work.

4. Results of the dissertation and specific contributions of its author

The results that were presented: development of models of the NV photoexcitation dynamics that were used to explain several experimental observations, demonstration of a microwave-free photoelectrical readout, and design and fabrication of diamond chips with electrode structures. The author presents his contribution at the end of each chapter. From what I understood, the author only contributed on the theoretical side, i.e. model the chip design and NV photoexcitation dynamics. All experimental work was done by others.

5. Relevance for the practice and development of the field of study Biomedical and Clinical Technology

While the field of quantum sensing using NV centers has begun to mature in the last decade in its scope and potential for applications, real-world use cases are only recently starting to take shape. Especially biochemical/-medical applications using NV-based sensing are still in their infancy. The work presented in this thesis is thus very timely regarding the exploration of what might be possible and has high potential.

6. Formal arrangement of the dissertation thesis and its language level

The formal arrangement of the thesis is fine and suits the topic. However, regarding the language level, a lot should be improved here. Apart from typos, which should be easy to fix with proper spell checkers, there are grammatical mistakes, wrong word order and sentence constructions that sometimes prevent understanding the actual content. Some examples:
Pg. 3: "Discrepancies between the spins ms=0 and ms=±1, can be further measured by elucidated through the generation of charge carriers, ..."
Pg. 5: "While for the readout techniques discussed above, diamond a slab integrated with a chip is used...."

Also figures should be checked, such as on pg.10, Fig.4, where type Ib is mislabeled as type Ia.

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7. Comments and final evaluation of the dissertation thesis.

Many simulations and experiments presented are performed as a function of laser power. In my mind, the actual important parameter however should be the intensity, i.e. power/area. Maybe I missed it but I couldn't find any hints for converting that anywhere, for instance what beam spot size is assumed in the simulations or used in experiments.

At some point, an acceptor level X is introduced. What could be a good way to find out what type of defect that is?

Somewhat related, it has been shown previously that electrons excited to the conduction band have good probability to recombine into neutral nitrogen (and not only positively-charged), thereby creating negatively-charged nitrogen (see PRB 84, 165202), a scenario that is not considered here explicitly. Would considering that change anything in the simulations?

Judging from the motivation, the work presented in this thesis is a first stepping stone for future work to bring NV-based sensing to biomedical applications, with ambitious goals like 'nanometric resolution' even being present in the thesis title. Considering that, a more detailed outlook for future work would be beneficial, i.e. what actually can realistically be achieved. For instance, I can see how imaging can be done using PL, and results are presented (although not how to get to 'nanometric resolution'), but what about photocurrent? How will one obtain spatial information? Or is that not a goal? It is not really clear to me.

Overall, the thesis is of high quality and I recommend its defense.

Mainz, 04.06.24

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