

## Summary of the changes

We would first like to thank the referees for the time they spent reviewing of our manuscripts.

[Résumer la position de ref et dire qu'ils ont raison meme si c'est pas vrai]

We agree with these remarks and have amended the manuscript in consequence to the best of our abilities.

## Referee 1

### Referee 1 report

This manuscript describes experiments showing mechanically detected cross-relaxation resonance of nitrogen-vacancy centers in diamond. The measurements are carried out on NV-centers hosted in nano-diamonds levitated in a Paul trap in vacuum. The authors motivate their work as a means of detecting paramagnetic defects without optical transitions, for investigating spin relaxation processes, and for cooling the motion of mechanical oscillators.

The work appears valid and the interpretation of the measurements and the accompanying analysis appears well-founded. Although the presentation is sometimes difficult to follow, the authors have included an exhaustive supplementary section spelling out the details of the experiment.

Nevertheless, the importance and broad interest of the work – requirements for publication in PRL – appear to this reader to fall below threshold. I am hard pressed to identify how this work, "substantially advance[s] a field, open[s] a significant new area of research, or solves[s]—or take[s] a crucial step toward solving—a critical outstanding problem". Cross-relaxation is a well-known phenomenon, also in NV centers. Although it appears not to have been studied in nano-diamonds nor to have been detected mechanically, it is not clear why these achievements solve an outstanding problem of interest.

The authors suggest in the conclusion that the scheme can be used to cool a nano-diamond in the Paul trap, though no such scheme is attempted in the paper. Also, no estimates are given as to how efficient this process would be and what the minimum achievable temperature would be. The second motivation offered is that this method could be used as a spectroscopic technique for sensing dipolar interaction between NV centers and spins that are not optically active. Again, no such demonstration experiment is presented, although it's not clear why this should not be possible. The final motivation given is for "bottom-up investigations of magnetism".

Unfortunately, from the description that follows this statement, it's hard to envision what, specifically, is intended.

For these reasons, it is my opinion that the authors should resubmit to PRB or a specialized journal of magnetic resonance.

## Our answer

Here is our answer to the points raised by Referee 1 :

Je sais pas trop quoi répondre sachant qu'il ne parle que de la conclusion. Est-ce qu'on change la conclu (la partie bottom-up magnetism, j'avoue que je vois pas bien ce que ça cible) ? Est-ce qu'on dit qu'on a pas réussi les applications ? Sur la partie détection de spin au moins je peux justifier : les seuls autres spins qu'on sache être présent ici c'est les P1 et non seulement c'est galère d'aligner précisément un champ mag sur la 111, en plus les effets de para/diamagnétisme des NV commencent à être significatifs à 500G.

## Referee 2

### Referee 2 report

In this work the authors investigate using dipolar interactions between NV centers to enhance magnetic torque and rotate electrically-charged, paramagnetic diamond nanocrystals that are levitated in an ion trap, through a mechanism similar to the Einstein-de-Haas effect for ferromagnets. An external magnetic field is tuned to produce resonant cross-relaxation, resulting in greater paramagnetism and spin torque. Prospects for using this approach to sense dipolar interactions between NV centers and spins that cannot be polarized optically are interesting. Using NV centers in levitating diamonds for emulating magnetism at the  $100 \mu_B$  level is another potentially useful application. I feel this work is novel and interesting however the paper requires clarifications before I can recommend publication.

Fig 3: the caption describes “a) and b) show the torque with and without cross-relaxation between NV centers respectively.” should this description be reversed? I thought b) was with CR.

The experimental setup figure in the supplemental material is showing one APD, but the figure in paper shows 2 detectors: APD1 and APD2. This should be consistent.

A related question: how is the actual angular motion determined and calibrated? I find the only explanation of how the angle is measured on pg 10 of the supplemental material “focus a small area of the speckle image onto an optical fiber and detect the photons transmitted through the fiber with the APD1. The detected signal is then highly sensitive to the particle position and orientation.”

How exactly is it sensitive and how is the calibration determined? This should be better explained in the main text as well as the Supplemental Material.

The work described in the supplemental material Fig. 3 regarding measuring the PL is done with a static micro diamond. How is the setup arranged as compared to the work in the Paul trap.

I was surprised there was not even more drift of the orientation of a trapped nanodiamond in the Paul trap. Can the authors account for the physics of the drift, in the SI a suggestion is made “The most likely origin of this drift is the loss of charges of the diamond due to photoionization by the laser, which changes the trapping conditions over time.” Can this somehow be made more quantitative? It seems like for better understanding and controlling the mechanical rotation for future applications this may be important to understand?

## **Our answer**

Here is our answer to the points raised by Referee 2 :

We have inverted the description of figure 3 in the main text to accurately describe the figure. (A FAIRE !j'ai pas le fichier à jour du main text)

We have changed the experimental setup figure in the supplementary material to be consistent with the one in the main text

La calibration de l'angle : Je pense qu'on peut rediriger vers le SI du nature de Tom (il trouve une précision de 0.3 mrad/racine(Hz)). Par contre je sais pas dans quel mesure on développe la réponse.

The static experiments have been performed in the same experimental setup, with the diamonds deposited on the trap electrode/antenna instead of levitating inside it.

Le drift : je vois pas trop quoi dire