Quantum Sensing of Insulator-to-Metal Transitions in a Mott Insulator

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Nitrogen vacancy (NV) centers, optically active atomic defects in diamond, have attracted tremendous interest for quantum sensing, network, and computing applications due to their excellent quantum coherence and remarkable versatility in a real, ambient environment. Taking advantage of these strengths, this paper reports on NV-based local sensing of the electrically driven insulator-to-metal transition (IMT) in a proximal Mott insulator. The resistive switching properties of both pristine and ion-irradiated VO₂ thin film devices are studied by performing optically detected NV electron spin resonance measurements. These measurements probe the local temperature and magnetic field in electrically biased VO₂ devices, which are in agreement with the global transport measurement results. In pristine devices, the electrically driven IMT proceeds through Joule heating up to the transition temperature while in ion-irradiated devices, the transition occurs nonthermally, well below the transition temperature. The results provide direct evidence for nonthermal electrically induced IMT in a Mott insulator, highlighting the significant opportunities offered by NV quantum sensors in exploring nanoscale thermal and electrical behaviors in Mott materials.

1. Introduction

As technology begins to reach the fundamental limitations of the von Neumann computing architectures, new paradigms are urgently required to improve the processing speed, data storage capacity, and energy efficiency for next-generation information technologies.[1] Among many potential contenders, neuromorphic computing[2–4] has attracted immense research interest due to its capability to mimic the highly interconnected structure of biological neural systems like the human brain.[5] Insulator-to-metal transitions (IMTs) in Mott materials featuring first-order, threshold firing type resistive switching behavior are directly relevant in this context due to their significant potential for implementing artificial spiking neurons in neuromorphic circuits.[6–10] Thus, great efforts have been devoted to understanding and controlling the mechanism for resistive switching in materials exhibiting an IMT.[11–15] Successful applications of IMTs to emergent neuromorphic computing platforms require advances in theory, material discovery, and equally importantly, a detailed knowledge of the local electrical and thermal properties of Mott materials down to the nanoscale regime.

Conventional research on IMTs has been mainly focused on global electrical transport measurements as well as structural characterizations, rendering limited information on the local properties of the studied materials. To address this challenge, here, we utilized nitrogen vacancy (NV) centers, optically active atomic defects in diamond that act as single-spin sensors[16] to perform local quantum sensing of the voltage-induced IMT in a prototypical Mott material: vanadium dioxide VO₂.[3,7,17] Notably, the measured magnetic fields generated by the VO₂ devices exhibit a characteristic step-like jump around a "critical" electric current, in agreement with the formation of conducting filaments during the voltage-induced IMT.[3,18] The temperature profile we observed for both pristine and ion-irradiated VO₂ films are explained by thermal[19] and nonthermal origins.[20] We expect that the presented NV-based quantum sensing platform can be extended naturally to other Mott insulators, offering a new perspective to reveal the local thermal and electrical behaviors in Mott-material-based neuromorphic devices.

The measurement platform and device structure used are illustrated in Figure 1a. We have grown 170 nm thick VO₂ films on Al₂O₃ (012) substrates by radio-frequency magnetron sputtering.[1] Two 125 nm thick Ti/Au electrodes were fabricated with a separation of 10 µm on top of a VO₂ film for electrical transport measurements. Patterned diamond nanobeams containing individually addressable NV centers[21] were transferred on top of the VO₂ film and positioned between the two Au electrical contacts as shown by a scanning electron microscopy image in Figure 1b. The diamond nanobeam has the shape of an equilateral triangular prism with dimensions of 500 nm × 500 nm × 10 µm. The NV-to-sample distance was estimated to...
be \approx 100 \text{ nm},^{[22]} ensuring sufficient thermal and field sensitivity. An Au stripline was fabricated next to the electrical contacts to provide microwave control of the NV spin states.\[^{[22,23]}\] The whole sample was mounted on a heating stage to allow for a precise adjustment of the base temperature. IMTs can be thermally and/or electrically triggered in the VO\textsubscript{2} device, accompanied by orders of magnitude decrease in electrical resistivity.\[^{[19,20,24]}\] The NV center positioned on top of the VO\textsubscript{2} film serves as a local probe of the temperature and magnetic field at the NV site. Taking advantage of their excellent quantum coherence and single-spin sensitivity,\[^{[16,25,26]}\] NV centers have been demonstrated to be a transformative tool in exploring the local magnetic, electric, and thermal features of a variety of materials with unprecedented sensitivity and spatial resolution.\[^{[25,27–30]}\] Figure 1c shows a confocal optical image of an NV center positioned on the symmetry axis of the two Au electrical contacts, demonstrating the single-spin addressability of our measurement system.

2. Results and Discussion

We first performed electrical transport measurements to characterize the temperature induced IMT in the VO\textsubscript{2} device. Figure 2a shows the resistance of the VO\textsubscript{2} device as a function of the base temperature. The blue and the red curves correspond to the heating and cooling branches, respectively. A resistance-temperature profile is shown in Figure 2a. When sweeping the current up, the filament shrinks and supports the metallic state, leading to a significant drop of the measured voltage as shown in Figure 2b. This is due to a metallic filament forming between the Au contacts which is visible by optical microscopy due to the change in optical constants (Figure 2c). Noting that the magnitudes of the critical currents and voltages of the electrically induced IMT depend on the base temperature of the device. The closer the base temperature is to \( T_c \), the smaller the electric power is required to activate the resistive switching as illustrated in Figure 2d. When the base temperature of the device is too far below \( T_c \), it is not possible to trigger voltage-induced IMTs in the VO\textsubscript{2} film without reaching excessively high voltages, where irreversible phase variations into other vanadium oxide compounds could occur.\[^{[24]}\] In the following NV measurements, the base temperature of the sample is set to be above 295 K to avoid irreversible damage to the VO\textsubscript{2} devices. The base temperature was maintained at a constant value below \( T_c \), while an electric current \( I_{dc} \) applied in the VO\textsubscript{2} channel was slowly varied and the voltage was simultaneously measured between the two Au contacts. At a certain critical current \( I_c \), a conducting filament stretching between the tips of the two Au contacts was formed in the VO\textsubscript{2} film, leading to a sudden and significant drop of the measured voltages as shown in Figure 2b.

Next, we demonstrate the NV center’s ability to accurately detect the local temperature and magnetic field environment of the VO\textsubscript{2} device during the voltage-induced IMTs. The negatively charged NV state has an \( S = 1 \) electron spin with a spin triplet ground state. Figure 3a shows the energy levels of an NV spin as a function of an external magnetic field \( (B_{//}) \) applied along the NV-axis. When \( B_{//} = 0 \), the \( m_s = \pm 1 \) states of the NV spin are degenerate with a characteristic NV zero-splitting frequency.

Figure 1. a) Schematic of a prepared NV-VO\textsubscript{2} device consisting of two Au electrodes and an Au microwave stripline fabricated on top of a 170 nm thick VO\textsubscript{2} thin film. A diamond nanobeam containing individually addressable NV centers is transferred on top of the VO\textsubscript{2} film to perform local thermal and field sensing of IMT. b) A scanning electron microscopy image showing a patterned diamond nanobeam situated between the Au electrical contacts. c) A photoluminescence image showing a diamond nanobeam containing a single NV spin positioned between the two the Au electrical contacts.

Figure 2. a) Resistance of a pristine VO\textsubscript{2} device measured as a function of the base temperature. Blue and red curves correspond to the heating and cooling branches, respectively. b) The voltage \( (V) \) measured as a function of applied current \( (I_{dc}) \) at a base temperature of 317 K. Blue and red curves correspond to increasing and decreasing current, respectively. When sweeping the current down, the filament shrinks and supports the metallic state with a lower critical current. c) An optical image showing the formation of a conducting filament (dark color) between the two Au contacts. d) Electric voltage \( (V) \) measured as a function of the applied current \( (I_{dc}) \) in the VO\textsubscript{2} device at three different base temperatures.
NV PL intensity was measured as a function of the microwave frequency $f$. When $f$ matches the NV ESR energies, the NV spin tends to flip from the $m_s = 0$ to the $m_s = \pm 1$ states, giving rise to two dips in the measured ESR spectrum. The Oersted field generated by the electric current flowing the VO$_2$ film can be obtained by the Zeeman splitting of the NV ESR frequencies

$$\Delta B_{||} = \frac{\pi f_+ - \pi f_-}{\gamma} - B_{||}$$

where $f_\pm$ correspond to the NV ESR frequencies of the $m_s = 0 \leftrightarrow \pm 1$ transitions and $\Delta B_{||}$ is the component of the Oersted field that is parallel to the NV-axis as illustrated in Figure 3b. The local temperature $T_I$ at the NV site can be extracted from the NV ESR measurements as follows: $T_I = \frac{f_+ + f_-}{2} - \frac{a}{b}$, where $a$ and $b$ are fitting parameters equal to $2.8983 \pm 0.002$ GHz and $-88.9 \pm 5.8$ kHz K$^{-1}$, respectively. These values are obtained by measuring $T_I$ as a function of the sample temperature (see the Supporting Information for details). Since the measurements were performed in a vacuum environment and due to the nanoscale proximity established between the NV center and the VO$_2$ sample, the NV center is well thermalized with its immediate vicinity in the device (see the Supporting Information for details) and the local temperature of the VO$_2$ sample can be measured. Figure 3c shows a typical NV ESR spectrum with $B_{||} = 700$ Oe at a base temperature of 295 K.

Figure 3d,e shows the extracted Oersted field $\Delta B_{||}$ and the local temperature $T_I$ as a function of the applied electric current $I_{dc}$ at three different base temperatures of 320, 325, and 330 K. For low electric currents $I_{dc} < I_c$, VO$_2$ is in a homogeneous semiconducting state and the electric current is sparsely distributed in the device, leading to a negligibly small Oersted field at the NV site. When the electric current reaches its critical value ($I_{dc} = I_c$), the IMT is electrically triggered, accompanied by the formation of a conducting filament in the VO$_2$ film. Since the electric current is then mainly concentrated in the conducting filament, the local current density and the Oersted field experienced by the NV center are significantly enhanced (Figure 3d). These sudden jumps in magnetic field correspond to switching events observed in the electrical transport measurements. The measured Oersted fields around the critical currents are in qualitative agreement with COMSOL simulations (see the Supporting Information for details). In contrast to the jump-type variation of the Oersted field, the measured local temperature $T_I$ exhibits a gradual increase as a function of $I_{dc}$. It is worth mentioning that $T_I$ measured at different base temperatures reaches a similar value ($\approx 335$ K) during resistive switching, demonstrating the thermal origin of the voltage-induced IMT observed in a pristine VO$_2$ film$^{[19,36]}$. At a sufficiently large electric current, Joule heating along the current path locally increases the temperature of the VO$_2$ film to the critical value ($\approx 335$ K) and triggers the formation of the conducting filaments.

In addition to Joule heating, it has been suggested that electric fields may also induce the IMT, but the origin of this effect is still debated$^{[20,37-43]}$. A number of reports have indicated that the IMT may be induced without reaching the IMT critical temperature$^{[12,13,36,44-47]}$. However, due to the inhomogeneous nature of the IMT and the need for simulations of the current and temperature distribution in the sample, direct evidence for...
the nonthermally induced IMT remains elusive. Local measurements of temperature in the nanoscale during the IMT are of great value to confirm the possibility of nonthermal switching. Next, we applied the NV-based quantum sensing platform to ion-irradiated VO$_2$ thin film devices to access the mechanism of nonthermally induced IMT\(^\text{[23]}\). We employed a focused ion beam to irradiate gallium ions onto the VO$_2$ film in a \(\approx 2\) \(\mu\)m wide region that connects the Au contacts. The gallium irradiation has interesting effects on the transport properties of the VO$_2$ thin film. First of all, the voltage-induced IMTs can be triggered at a much lower base temperature with reduced current/voltage/power as shown in Figure 4a. However, the resistance-temperature characteristics of the ion-irradiated VO$_2$ device remain largely the same, showing a similar \(T_c\) as in the pristine sample (Figure 4b). Figure 4c shows the local temperature extracted from the NV ESR measurements as a function of \(I_{dc}\). In stark contrast to the pristine VO$_2$ sample, the local temperature measured at the critical current barely changes from the base temperature which can be up to 35 K lower than \(T_c\). It is important to note that the NV center is situated on the symmetry axis between the two contacts and is in contact with the conducting filament formed in the ion-irradiated VO$_2$ device (see the Supporting Information for details). The absence of substantial heating both before and after the switching process demonstrates the nonthermal origin of the electrically induced IMT in the ion-irradiated VO$_2$ sample\(^{[20]}\).

Our results are consistent with a previous study showing indirect evidence for nonthermal switching and support field-assisted carrier generation as the switching mechanism (see the Supporting Information for details)\(^{[20,48,49]}\). With a sufficiently large electric field, in-gap states created by ion beam irradiation could be electrically excited, emitting charge into the conduction band, as illustrated in Figure 4d. This increases the number of free carriers and causes the collapse of the Mott insulator state through a doping-driven IMT\(^{[20]}\). In comparison to thermally induced resistive switching, we highlight that the critical currents and energy dissipations are significantly reduced in the doping-driven IMT, offering significant advantages for developing energy-efficient neuromorphic circuits in a broad temperature range.

### 3. Conclusions

In summary, we have demonstrated NV centers as a sensitive local probe of the thermally and nonthermally induced IMTs in VO$_2$ devices. By measuring the local temperature and the magnetic field environment via the optically detected NV electron spin resonances, the underlying electrical phase transitions in proximal VO$_2$ devices could be accessed in a nonperturbative way at the nanoscale. This technique allowed us to obtain direct evidence for a nonthermally induced electrical IMT in a Mott insulator. The findings also have important implications for our understanding of the physics of Mott insulators and their applications. By employing patterned diamond nanostructures with shallowly implanted NV centers\(^{[50–52]}\), we expect that the local resolution of such NV quantum sensing platform could potentially reach the few-nanometer length scale, offering new opportunities to reveal the electrical and thermal behaviors in Mott insulators and many other quantum materials. The demonstrated coupling between NV centers and Mott insulators may also find applications in developing next-generation, hybrid neuromorphic devices.

### 4. Experimental Section

**Materials and Device Fabrication:** The 170 nm thick VO$_2$ films used in this work were deposited by reactive radio frequency magnetron sputtering on (012)-oriented Al$_2$O$_3$ substrates. The growth was done in a 4 mTorr Ar:O$_2$ (92–8%) atmosphere and at a substrate temperature of 470°C. After the growth, the sample was cooled down at a rate of 12 °C min$^{-1}$. For electrical measurements and microwave pumping, Ti (15 nm)/Au (110 nm) electrodes and a stripline were fabricated on top of pristine VO$_2$ films using standard photolithography and e-beam evaporation techniques. After the electrode fabrication, some devices were irradiated using a focused Ga-ion beam. The device region was irradiated with \(6.2 \times 10^{15}\) ions cm$^{-2}$ by rastering a 30 keV focused ion beam with 50% overlap between successive raster locations to ensure a uniform dose in the irradiated region.
The width of the irradiated region was ≈2 μm and the length extended just between the Ti/Au electrodes to minimize damage to the electrodes.

Patterened diamond nanobeams containing individually addressable NV centers were picked up and transferred onto the VO₂ thin films using a tungsten tip performed under a micromechanical transfer stage. Nanobeams were fabricated by a combination of top-down etching and angled etching processes. Acid cleaning was performed before and after the fabrication processes to ensure a pristine diamond surface. The angle between the NV-axis and the surface normal was 46.1° and 68.4° for the two NV centers positioned on the pristine and ion-irradiated VO₂ devices, respectively. The direction of the NV-axis was determined by magnetic field alignment. The NV photoluminescence was experimentally monitored as a function of the position of a permanent magnet. The direction and magnitude of the magnetic field generated by the permanent magnet could be precisely calculated based on its dimensions and moment, which was further confirmed by NV ESR measurements. When the NV-axis was parallel with the direction of the external magnetic field, NV centers emitted maximum photoluminescence which could be optically identified.

**NV Electron Spin Resonance Measurements**: NV ESR measurements were performed by a scanning confocal microscope. A continuous green laser was applied at the NV site to constantly initialize the NV spin to the m∥ = 0 state. The laser power was fixed at 0.2 mW. Microwave signals were delivered to microwave switches; a microwave combiner, an amplifier, and finally were sent to the on-chip Au stripe line to excite the NV ESR. The input microwave power was set to be ~30 dBm before amplification. The microwave amplifier gain was about 50 dB in the frequency range of interest. NV ESR was optically detected by monitoring the photoluminescence as a function of microwave frequency through an avalanche photodiode. The trigger pulses to the optical modulator and photon counting were generated by a programmable pulse generator. The external magnetic field B∥ was generated by a cylindrical NdFeB permanent magnet attached on a scanning stage.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

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

**Keywords**

insulator-metal transition, Mott insulators, neuromorphic engineering, nitrogen vacancy magnetometry, quantum sensing

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[1] J. von Neumann, IEEE. Hist. Comput. 1993, 15, 27.
[2] D. Monroe, Commun. ACM 2014, 57, 13.
[3] J. del Valle, P. Salev, F. Tesler, N. M. Vargas, Y. Kalcheim, P. Wang, J. Trastoy, M.-H. Lee, G. Cassabian, J. G. Ramirez, M. J. Rozenberg, I. K. Schuller, Nature 2019, 569, 388.
[4] H. T. Zhang, P. Panda, J. Lin, Y. Kalcheim, K. Wang, J. W. Freeland, D. D. Fong, S. Priya, I. K. Schuller, S. K. R. Sankaranarayanan, K. Roy, S. Ramanathan, Appl. Phys. Rev. 2020, 7, 011309.
[5] C. Mead, Proc. IEEE 1990, 78, 1629.
[6] M. Imada, A. Fujimori, Y. Tokura, Rev. Mod. Phys. 1998, 70, 1039.
[7] J. H. Park, J. M. Coy, T. S. Kasirga, C. Huang, Z. Fei, S. Hunter, D. H. Cobden, Nature 2013, 500, 431.
[8] P. Stoliar, J. Tranchant, B. Corraze, E. Janod, M.-P. Besland, F. Tesler, M. Rozenberg, L. Cario, Adv. Funct. Mater. 2017, 27, 1604740.
[9] J. H. Ngai, F. J. Walker, C. H. Ahn, Annu. Rev. Mater. Res. 2014, 44, 1.
[10] T. M. Dao, P. S. Mondal, Y. Takamura, E. Arenholz, J. Lee, Appl. Phys. Lett. 2011, 99, 121111.
[11] J. S. Brockman, L. Gao, B. Hughes, C. T. Rettner, M. G. Samant, K. P. Roche, S. S. P. Parkin, Nat. Nanotechnol. 2014, 9, 453.
[12] G. Stefanovich, A. Pergament, D. Stefanovich, J. Phys.: Condens. Matter 2000, 12, 8837.
[13] P. Diener, E. Janod, B. Corraze, M. Querré, C. Adda, M. Guilloux-Viry, S. Cordier, A. Camjayi, M. Rozenberg, M. P. Besland, L. Cario, Phys. Rev. Lett. 2018, 121, 16601.
[14] D. Li, A. A. Sharma, D. K. Gala, N. Shukla, H. Paik, S. Datta, D. G. Schlom, J. A. Bain, M. Skowronski, ACS Appl. Mater. Interfaces 2016, 8, 12908.
[15] F. Giorgianni, J. Sakai, S. Lupi, Nat. Commun. 2019, 10, 1159.
[16] L. Rondin, J.-P. Tetienne, T. Hingant, J.-F. Roch, P. Maletinsky, V. Jacques, Rep. Prog. Phys. 2014, 77, 056503.
[17] D. Lee, B. Chung, Y. Shi, G.-Y. Kim, N. Campbell, F. Xue, K. Song, S.-Y. Choi, J. P. Podkaminer, T. H. Kim, P. J. Ryan, J.-W. Kim, T. R. Paudel, J.-H. Kang, J. W. Spinuzzi, D. A. Tenne, E. Y. Tsybulya, M. S. Rzchowski, L. Q. Chen, J. Lee, C. B. Eom, Science 2018, 362, 1037.
[18] A. Mansigh, R. Singht, J. Phys. C: Solid State Phys. 1980, 13, 5725.
[19] A. Zimmers, L. Aigouy, M. Mortier, A. Sharoni, S. Wang, K. C. West, J. G. Ramirez, I. K. Schuller, Phys. Rev. Lett. 2013, 110, 056601.
[20] Y. Kalcheim, A. Camjayi, J. del Valle, P. Salev, M. Rozenberg, I. K. Schuller, Nat. Commun. 2020, 11, 2985.
[21] M. J. Burek, N. P. de Leon, B. J. Shields, D. J. M. Hausmann, Y. Chu, Q. Quan, A. S. Zibrov, H. Park, M. D. Lukin, M. Lončar, Nano Lett. 2012, 12, 6084.
[22] C. H. Du, T. van der Sar, T. X. Zhou, P. Upadhyaya, F. Casola, H. Zhang, M. C. Onbasli, C. A. Ross, R. L. Walsworth, Y. Tserkovnyak, A. Yacoby, Nature 2017, 537, 195.
[23] G. D. Fuchs, V. V. Dobrovitski, D. M. Toyli, F. J. Heremans, D. D. Awschalom, Science 2009, 326, 1520.
[24] J. Del Valle, Y. Kalcheim, J. Trastoy, A. Charnukha, D. N. Basov, I. K. Schuller, Phys. Rev. Appl. 2017, 8, 054041.
[25] M. S. Grinolds, M. Warner, K. De Greve, Y. Dovzhenko, L. Thiel, R. L. Walsworth, S. Hong, P. Maletinsky, A. Yacoby, Nat. Nanotechnol. 2014, 9, 279.
[26] G. Balasubramanian, I. V. Chan, M. Kolesov, Al-Hmoud, J. Tsiler, C. Shin, C. Kim, A. Wojcik, P. R. Hemmer, A. Krueger, T. Hanke, A. Leitenstorfer, R. Bratschitsch, F. Jelezko, J. Wrachtrup, Nature 2008, 455, 648.
[27] G. Kucsko, P. C. Maurer, N. Y. Yao, M. Kubo, H. J. Noh, P. K. Lo, H. Park, M. D. Lukin, Nature 2013, 500, 54.
[28] D. M. Toyli, C. F. de las Casas, D. J. Christie, V. V. Dobrovitski, D. D. Awschalom, *Proc. Natl. Acad. Sci. USA* **2013**, *110*, 8417.

[29] F. Casola, T. van der Sar, A. Yacoby, *Nat. Rev. Mater.* **2018**, *3*, 17088.

[30] A. Laraoui, H. Aycock-Rizzo, Y. Gao, X. Lu, E. Riedo, C. A. Meriles, *Nat. Commun.* **2015**, *6*, 8954.

[31] M. M. Qazilbash, M. Breihl, B.-G. Chae, P.-C. Ho, O. Andreev, B.-J. Kim, S. J. Yun, A. V. Balatsky, M. B. Maple, F. Keilmann, H.-T. Kim, D. N. Basov, *Science* **2007**, *318*, 1750.

[32] E. Lee-Wong, R. L. Xue, F. Y. Ye, A. Kreisel, T. van der Sar, A. Yacoby, C. H. R. Du, *Nano Lett.* **2020**, *20*, 3284.

[33] P. Neumann, I. Jakobi, F. Dolde, C. Burk, R. Reuter, G. Waldherr, J. Honert, T. Wolf, A. Brunner, J. H. Shim, D. Suter, H. Sumiya, J. Isoya, J. Wrachtrup, *Nano Lett.* **2013**, *13*, 2738.

[34] V. M. Acosta, E. Bauch, M. P. Ledbetter, A. Waxman, L. S. Bouchard, D. Budker, *Phys. Rev. Lett.* **2010**, *104*, 070801.

[35] N. Wang, W.-H. L. G.-Q. Liu, H. Zeng, X. Feng, S.-H. Li, F. Dolde, H. Fedder, J. Wrachtrup, X.-D. Cui, S. Yang, Q. Li, R.-B. Liu, *Phys. Rev. X* **2018**, *8*, 011042.

[36] I. Valmianski, P. Y. Wang, S. Wang, J. Gabriel Ramirez, S. Guénon, I. K. Schuller, *Phys. Rev. B* **2018**, *98*, 195144.

[37] T. Oka, R. Arita, H. Aoki, *Phys. Rev. Lett.* **2003**, *91*, 66406.

[38] H. Yamakawa, T. Miyamoto, T. Morimoto, T. Terashige, H. Yada, N. Kida, M. Suda, H. M. Yamamoto, R. Kato, K. Miyagawa, K. Kanoda, H. Okamoto, *Nat. Mater.* **2017**, *16*, 110.

[39] B. Mayer, C. Schmidt, A. Grupp, J. Bühler, J. Oelmann, R. E. Marvel, R. F. Haglund, Jr., T. Oka, D. Brida, A. Leitenstorfer, A. Pashkin, *Phys. Rev. B* **2015**, *91*, 235113.

[40] T. Oka, H. Aoki, *Phys. Rev. Lett.* **2005**, *95*, 137601.

[41] B. Wu, A. Zimmers, H. Aubin, R. Ghosh, Y. Liu, R. Lopez, *Phys. Rev. B* **2011**, *84*, 241410.

[42] W.-R. Lee, K. Park, *Phys. Rev. B* **2014**, *89*, 205126.

[43] N. Sugimoto, S. Onoda, R. Nagaosa, *Phys. Rev. B* **2008**, *78*, 155104.

[44] P. Stoliar, M. Rozenberg, E. Janod, B. Corraze, J. Tranchant, L. Cario, *Phys. Rev. B* **2014**, *90*, 45146.

[45] E. Janod, J. Tranchant, B. Corraze, M. Querré, P. Stoliar, M. Rozenberg, T. Cren, D. Roditchev, V. T. Phuoc, M.-P. Besland, L. Cario, *Adv. Funct. Mater.* **2015**, *25*, 6287.

[46] P. Stoliar, L. Cario, E. Janod, B. Corraze, C. Guillot-Deudon, S. Salmon-Bourmand, V. Guiot, J. Tranchant, M. Rozenberg, *Adv. Mater.* **2013**, *25*, 3222.

[47] G. Gopalakrishnan, D. Ruzmetov, S. Ramanathan, *J. Mater. Sci.* **2009**, *44*, 5345.

[48] S. D. Ganichev, E. Ziemann, W. Prettl, I. N. Yassievich, A. A. Istratov, E. R. Weber, *Phys. Rev. B* **2020**, *61*, 10361.

[49] J. Frenkel, *Phys. Rev.* **1938**, *54*, 647.

[50] L. Thiel, D. Rohner, M. Ganzhorn, P. Appel, E. Neu, B. Müller, R. Kleiner, D. Koelle, P. Maletinsky, *Nat. Nanotechnol.* **2016**, *11*, 677.

[51] M. Pelliccione, A. Jenkins, P. Ovartchiyapong, C. Reetz, E. Emmanouilidou, N. Ni, A. C. Bleszynski Jayich, *Nat. Nanotechnol.* **2016**, *11*, 700.

[52] S. J. Devience, L. M. Pham, I. Lovchinsky, A. O. Sushkov, N. Bar-Gill, C. Belthangady, F. Casola, M. Corbett, H. Zhang, M. Lukin, H. Park, A. Yacoby, R. L. Walsworth, *Nat. Nanotechnol.* **2015**, *10*, 129.