

Materials for Quantum Technology

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**PERSPECTIVE**

Strained diamond for quantum sensing applications

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Abstract

Apart from being an extraordinary optical and electronic material, diamond has also found applications in quantum mechanics especially in quantum sensing with the discovery and research development of various color centers. Elastic strain engineering (ESE), as a powerful modulation method, can tune the quantum properties and improve the performance of diamond quantum sensors. In recent years, deep ESE (DESE, when $>5\%$ elastic strain, or $>\sigma_{\text{ideal}}/2$ is achieved) has been realized in micro/nano-fabricated diamond and shows a great potential for tuning the quantum mechanical properties of diamond substantially. In this perspective, we briefly review the quantum properties of diamond and some of the corresponding sensing applications carried out with ESE, and look at how DESE could be applied for further tuning the quantum sensing properties of diamond with desired applications and what the critical challenges are.

1. Introduction

Strain engineering is a strategy to tune the physical and chemical properties of a material such as electronic, magnetic, and catalytic properties by applying strain which changes the lattice parameters and shape [1, 2]. Very often, the strain value does not need to be very high, but can change the material properties fundamentally and improve the device performance dramatically. For example, a small strain of $\sim 1\%$ can enhance the carrier mobility of Si-based field-effect-transistor by more than 50% [3, 4], thus improve the device performance to a greater extent. The implementation of strain engineering relies on the preparation of material with small size and less defects, that is, with the reduction of sample size, the probability of the appearance of internal defects is also reduced, making the material properties more uniform and closer to the theoretical level. Therefore, large strains are more likely to be achieved in nano- and single-crystalline material, which is also known as 'smaller is stronger' [5].

Strain can modulate the quantum mechanical properties of materials by changing energy or breaking symmetry. Firstly, strain changes the interatomic distance and interaction among different atomic orbitals, and modulates the relative position of energy levels and spin states of electrons. For example, strain tunes the energy levels and increase the orbital splitting of the silicon-vacancy (SiV) centers of diamond. By doing so, the spin coherence of SiV center can be improved [6]. Secondly, since the symmetry of a material is closely connected to its energy degeneracy, when strain breaks a specific symmetry of a system, some spin transitions which are not possible without strain can be enabled [7, 8], and more spin manipulation will become possible, such as the spin-spin interaction change of nitrogen-vacancy (NV)⁻ [9] and the generation of spin relaxation path of the transition-metal dichalcogenides [10] through strain-induced symmetry breaking. In recent years, strain engineering is appearing as an effective method to modulate the quantum mechanical properties of materials, such as modulating the quantum ferroelectric polar and thus the superconducting phase and superconducting transition temperature of materials [11, 12], which has been

accelerating the research of high temperature superconductors [13–15]. Moreover, strain can improve the quantum emission of materials such as hexagonal boron nitride film [16] and transition metal dichalcogenides [17–20] and extend their quantum photonic applications.

Diamond is a promising candidate for quantum mechanics and quantum technology applications such as quantum communication [21, 22], quantum computing [23–25] and quantum electromagnetic sensing [26–30]. With the development of diamond synthesis and nano-fabrication technologies, Elastic strain engineering (ESE) and DESE have been achieved in recently years [31, 32], which makes strain engineering of diamond a hot topic. In this paper, we briefly review the quantum mechanics of diamond and look forward to how ESE and DESE may accelerate the development of diamond quantum mechanics and extend its quantum sensing applications.

2. Color centers and quantum applications of diamond

Pure diamond is composed entirely of carbon and is optically transparent from ultraviolet to the far infrared. However, when diamond is doped by other elements (nitrogen, silicon, boron etc.), point defects form. Optically active point defects are termed color centers [33], of which many have special quantum features. Up to now, hundreds of color centers have been studied experimentally or theoretically [34]. Due to the rigid lattice and chemical stability of diamond, these quantum features can be well protected, even under extreme conditions [34].

Among those color centers, the most widely studied one is NV center, which contains a substitutional nitrogen atom and a vacancy at adjacent site. NV centers can be prepared via nitrogen implantation, irradiation of nitrogen-doped diamond with electron beam to create vacancies and a subsequent high-temperature annealing to combine the vacancies and nitrogen atoms into high-density NV centers, or vacancy creation and NV center formation via ultrahigh temperature annealing [34, 35]. The energy levels of a NV center at room temperature are shown schematically in figure 1(a), with the $m_s = 0$ and $m_s = 1$ states separated by the zero-field splitting parameter (D), which can be affected almost linearly by external strain [36]. NV center can exist in various charged states, i.e. negative (NV^-) or neutral charge (NV^0) states. The zero-phonon line (ZPL) of NV^- and NV^0 center are at 637 nm (1.945 eV) and 575 nm (2.156 eV), respectively and the NV^- center has a broad band from 650–750 nm [37–40]. The spin state of NV^- is sensitive to magnetic and electric field change and mechanical strain, and such change will reflect in its fluorescence signals [41–43]. Moreover, NV^- center has a long spin coherence time (T_2 , the time duration that a qubit maintains its quantum state), which can reach a few milliseconds using the Hahn echo in ambient conditions [44, 45], and can be extended to one second or over using dynamical decoupling under cryogenic conditions [46, 47]. Such a long coherence time facilitates NV[−] center to be initialized, manipulated and read out, often by using the optically detected magnetic resonance (ODMR). Therefore, diamond with NV center is an excellent material for quantum memory (figure 1(b)) [48–50], magnetic and electric field sensing (figures 1(c) and (d)) [43, 51–56], and computing [23, 25]. For example, as shown in figure 1(b), in a quantum memory protocol, a horizontally polarized photon with a wavelength of 723 nm (green light) is written with a vertically polarized write pulse with a wavelength of 800 nm (red). After a delay of τ , a horizontal read pulse recalls a vertical phonon. Although the spin coherence of NV center can also be affected by environmental sources of noise and disorder such as temperature, some strategies such as the local manipulation of the electron spin bath has been proposed to enhance the coherence and help read out signals [57, 58].

In addition to NV center, other color centers such as SiV, germanium-vacancy (GeV) and tin-vacancy (SnV) have also been investigated and found potential applications in quantum sensing. In the SiV structure, the Si occupies the center of two adjacent vacancies. SiV is a well-studied color center only second to NV center. Compared with the NV center, SiV center has a higher proportion (~70%) of its emission in ZPL (at 738 nm) and therefore allows a sharp ZPL to be detected, making SiV center advantageous in labeling [34, 59, 60], quantum communication [61] and quantum repeater [62]. Unfortunately, the coherence time of SiV center is too short (under no strain condition) and coherence spin control can also be realized by cooling down to low temperature [62]. GeV and SnV centers have geometric structures similar with SiV (that is, both Ge or Sn takes the place between two adjacent vacancies), as well as similar optical properties [63], but SnV has a longer spin coherence time than SiV and GeV at higher temperatures [64, 65]. Nevertheless, due to the large atomic size of Si, Ge and Sn, their correspondent color centers will bring more lattice distortion than the NV center does.

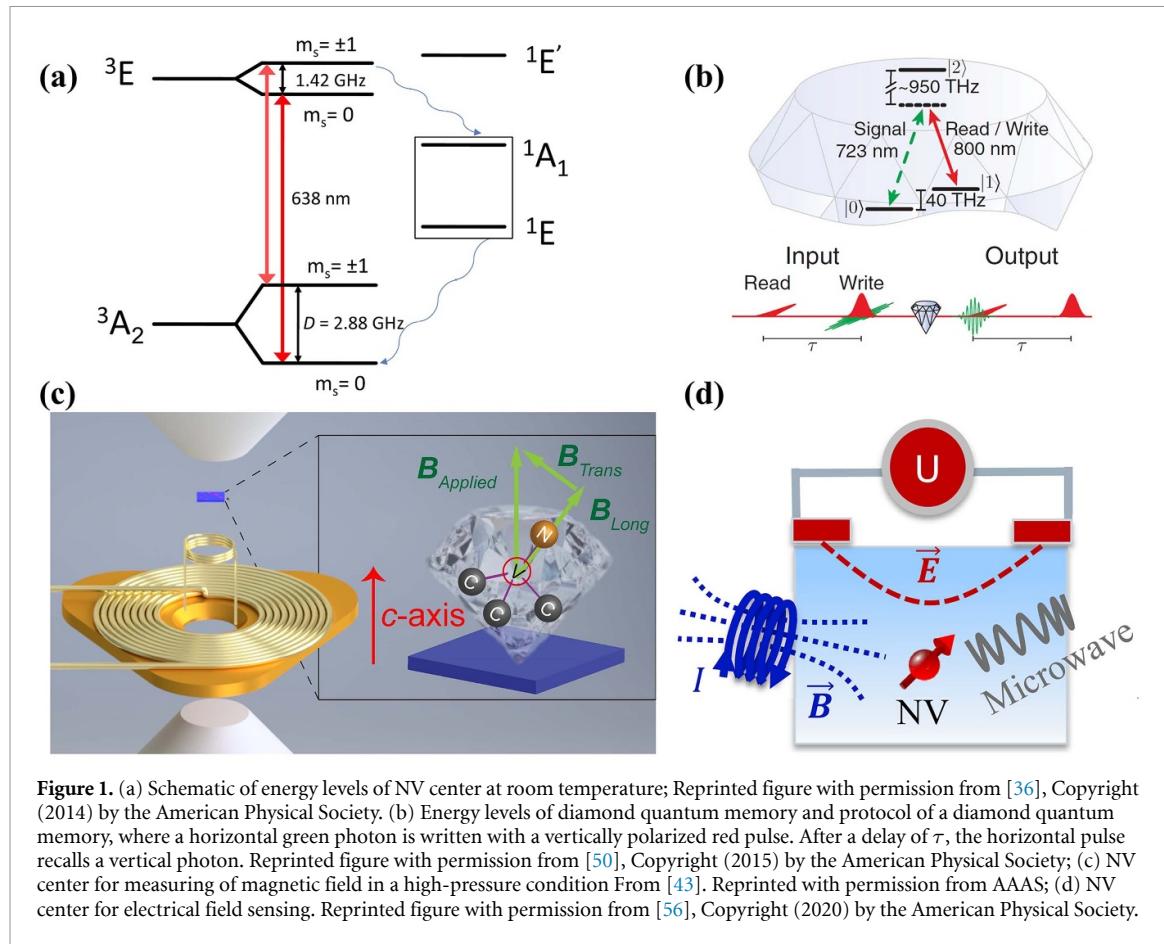


Figure 1. (a) Schematic of energy levels of NV center at room temperature; Reprinted figure with permission from [36], Copyright (2014) by the American Physical Society. (b) Energy levels of diamond quantum memory and protocol of a diamond quantum memory, where a horizontal green photon is written with a vertically polarized red pulse. After a delay of τ , the horizontal pulse recalls a vertical photon. Reprinted figure with permission from [50], Copyright (2015) by the American Physical Society; (c) NV center for measuring of magnetic field in a high-pressure condition From [43]. Reprinted with permission from AAAS; (d) NV center for electrical field sensing. Reprinted figure with permission from [56], Copyright (2020) by the American Physical Society.

3. Strain engineered quantum properties of diamond

Naturally, the orientations of NV centers (i.e. the direction NV axis is parallel with) are random, which would greatly compromise the device performance in quantum computing and sensing [69]. The alignment of NV centers can be achieved by carefully adjusting the parameters of the diamond growth during chemical vapor deposition (CVD) process, and of nitrogen implantation and high-temperature annealing [69–71]. ESE is a potentially alternative approach to modulate the atomic structure and align the orientations of the color centers. First principles calculations predicted that applying strains along some selected directions would make these orientations of the NV centers more energetically favorable over others, e.g. a compressive biaxial strain of 2% perpendicular to [111] direction or a small scale of tensile strain along [111] in high-temperature environment could make $\sim 90\%$ of NV centers align along the [111] directions (figure 2(a)) [66]. Such strategy could be employed to modulate the qubit coupling of NV centers already formed in diamond in order to make them ideal for quantum sensing and computing [72]. Moreover, diamond with well-aligned NV centers is also an excellent candidate for stress imaging [73] and high-accuracy magnetometry [74, 75]. Therefore, straining engineering can be employed as part of the post-processing treatments for the geometry optimization of the NV color center.

Strain often inevitably exists from intrinsic defects and can be caused by external stress as well. In some cases, strains are unwanted since they bring extra broadening of optical and spin spectra of NV centers [76]. But strain can be a powerful tool if used properly. For example, strain can modulate the orbital states and spin levels of NV center by shifting the spin states or inducing state mixing, such as the global shift of excited state, as shown in figure 2(b) [67, 77], or the coupling between the $|m_s = \pm 1$, which is an important effect of strain engineering tuning the quantum properties of diamond. For the NV⁺ center, the ZPL is highly dependent on strain field, which can split the 3E fine structure into two branches [40]. Based on these effects, strain susceptibilities can be obtained in a strain sensor for plotting the strain profile, which may find applications in hybrid quantum sensing devices such as nanomechanical resonators [78–81]. Since strain can shift the optical transition frequency of each electron spin, a strain field can be introduced for individually tuning the transition frequencies of NV centers embedded in a diamond cantilever, which enables manipulation of individual qubits and help to achieve high-fidelity quantum operations [82]. In addition,

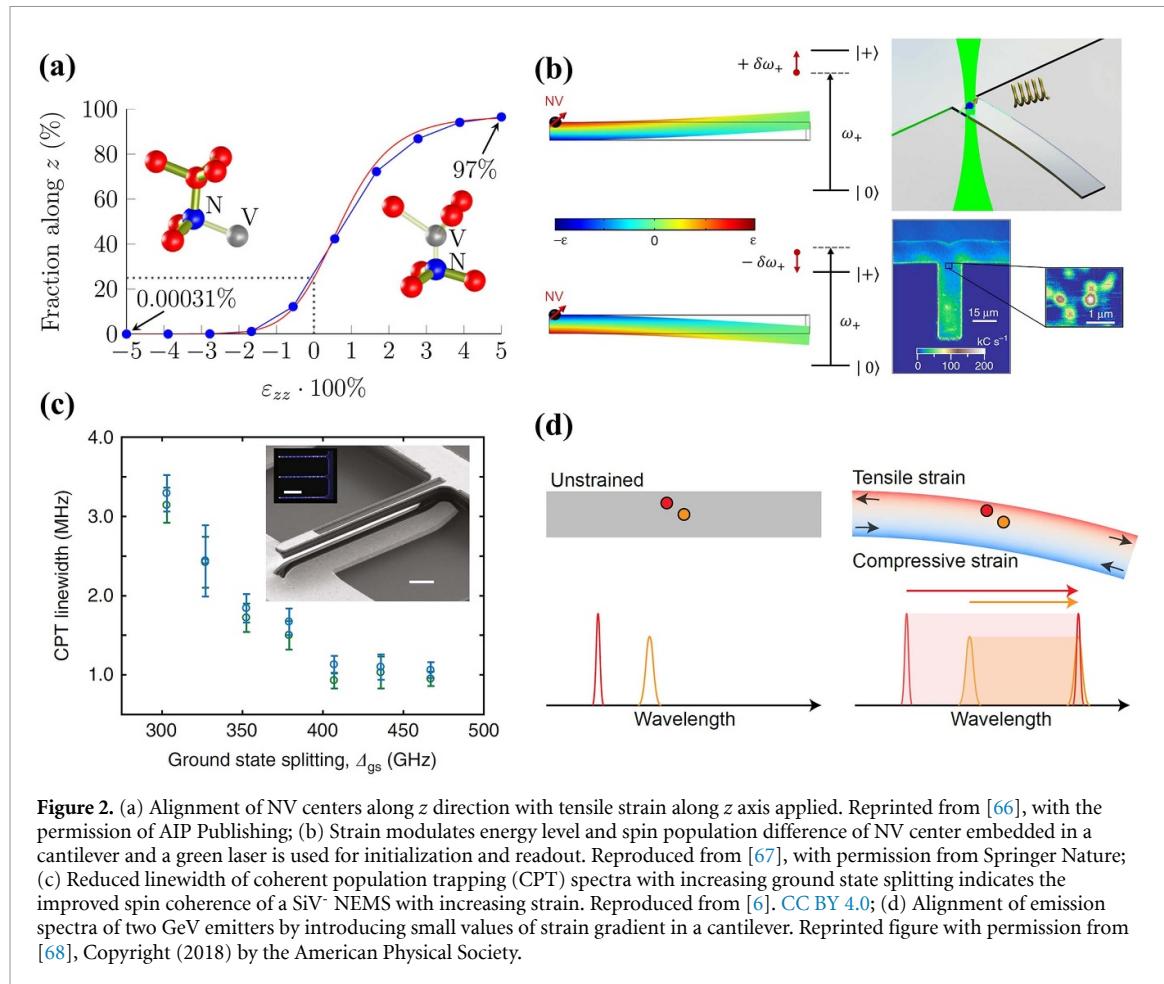
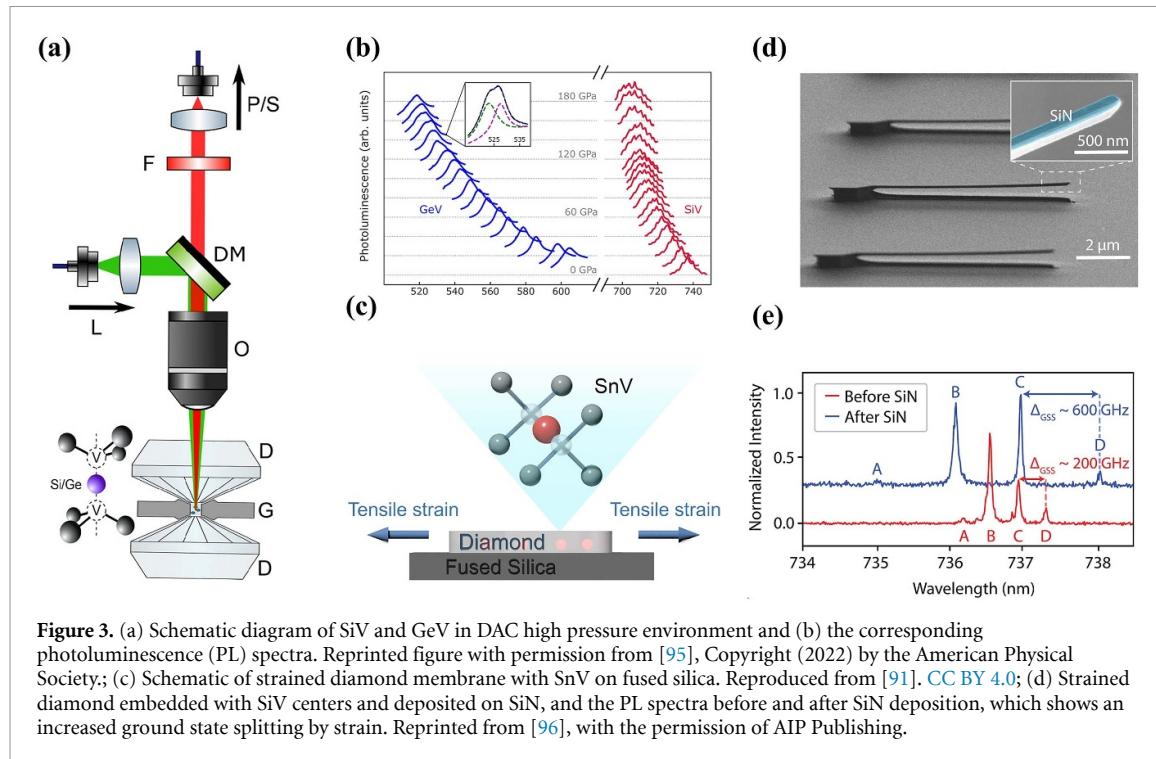


Figure 2. (a) Alignment of NV centers along z direction with tensile strain along z axis applied. Reprinted from [66], with the permission of AIP Publishing; (b) Strain modulates energy level and spin population difference of NV center embedded in a cantilever and a green laser is used for initialization and readout. Reproduced from [67], with permission from Springer Nature; (c) Reduced linewidth of coherent population trapping (CPT) spectra with increasing ground state splitting indicates the improved spin coherence of a SiV⁻ NEMS with increasing strain. Reproduced from [6]. CC BY 4.0; (d) Alignment of emission spectra of two GeV emitters by introducing small values of strain gradient in a cantilever. Reprinted figure with permission from [68], Copyright (2018) by the American Physical Society.

strain effect can also be used for tuning the nuclear spins in diamond lattice such as nitrogen or C¹³ atoms, which are useful for qubit registers and sensors [83, 84]. Since strain influences the distance and distribution of NV centers, which is strongly related to ODMR spectra, applying straining can be a way to facilitate the readout of nuclear spin. Moreover, by coupling the surface acoustic wave to the NV spin states through a dark state, it is possible to tune the NV spin states while without populating the excited state, therefore avoiding the unwanted decoherence [85]. Since strain fields can enhance the spin coherence of NV center, strain engineering could be employed together with electrical field tuning, which may bring noise and decoherence [86]. In addition, the coupling between strain and the NV spin could be used to facilitate the coherent spin-phonon interactions, which are important for quantum control and long-range spin-spin interactions [87, 88]. Apart from applying mechanical vibrations, using diamond anvil cell (DAC) is another way to introduce strain into diamond. In this way, compressive strain decreases the distance between atomic orbitals and makes the localized electrostatic potential of NV centers deeper. As a result, the electron density becomes more concentrated in the inner neighbor shells of the NV centers, which favors the spin-spin interaction [89].

In addition to the modulation of the quantum properties of NV center, strain engineering can also tune the properties and improve the performance of other color centers. For example, the electronic level structure of SiV⁻ center in a nano-electro-mechanical system (NEMS) can be modulated by applying static strain, which can be used for tuning the optical and spin transition frequencies [90]. Moreover, the thermal-induced quantum decoherence of SiV⁻ center in a NEMS can be effectively suppressed by applying a slight static strain (as shown in figure 2(c)), which provides a feasible way to control the phonon-induced decoherence of color centers in quantum emitters without lowering the operating temperature [6]. Similarly, static tensile strain, even of a small scale (0.05%–0.1%), can suppress the electron-phonon interaction of SnV in diamond at elevated temperatures effectively and improve the coherence time considerably [91]. For multiple GeV centers in a diamond cantilever, applying a strain gradient can tune the emission spectra of these GeV centers and achieve spectral alignment (as shown in figure 2(d)), which can promote the development of spectrally identical quantum emitters.



4. Deep strain engineering

Although strain engineering has been considered as a potential approach to modulate the quantum mechanical properties of material, in most cases, the strains applied are of small values (<1%) [1, 5]. Due to the unavoidable existence of defects (vacancies, interstitials, dislocations, cracks etc.), the mechanical performance of a material is often compromised such that it can hardly reach the ideal level. With the reduction of the material size to micro and even nano scale, the probability of the appearance of these defects can be reduced to lower level and the mechanical performance will be improved substantially. For example, it has been demonstrated that high-quality silicon nanowires can be stretched elastically above 10%, which is more than half of its theoretical limit (17%–20%) [92]. Accordingly, first principles calculations predicted an indirect to direct bandgap transition and a metallization transformation with ultra large strain of Si [93, 94], indicating the great potential of DESE, and one may think about what DESE can do for further tuning the optical, electronic and even quantum mechanical properties of more materials.

For diamond, due to its high brittleness, it is extremely difficult to achieve high elastic strain experimentally [97]. One of the most straightforward methods is to use DAC to generate ultrahigh static pressure. For example, with applying hydrostatic pressure of up to 180 GPa, large deformation of diamond can be obtained and blueshifts of ZPL of SiV and GeV can be observed and the shift becomes slower at high pressures, as shown in figures 3(a) and (b) [95]. Such a blue-shifting trend can also be observed in NV center under DAC [98]. Such work should be meaningful for carrying out quantum sensing at ultrahigh pressures. However, strain values cannot be measured with such a configuration and it is hard to guarantee a well uniform strain or to realize a controllable strain gradient. Therefore, the application of DAC tuning the quantum sensing properties is limited.

With the development of diamond synthesis and nanofabrication technology, high-quality single crystal diamond nano needles and bridge structures can be prepared by CVD followed by reactive/focused ion beam etching [31, 32, 99]. It was also found that diamond structure of such small size can sustain much higher elasticity compared to their bulk counterparts. For diamond nanoneedles, the highest elastic tensile strain reported was 13.4% and the compressive strain was even higher [99]. Moreover, diamond nano bridge structures exhibited a uniform elastic tensile strain of up to 9% and such structure may be applied for electronic, photonic and quantum devices [31]. Bandgap reduction and indirect to direct bandgap transformation were predicted by first principles calculations. Compared with indirect bandgap material, the direct bandgap materials can emit photons with higher efficiency, therefore, the strain-induced evolution of bandgap from indirect to direct makes DESE a potential approach to apply diamond to optoelectronic devices, such as laser diodes. Moreover, considering the deep-strain-induced bandgap reduction and indirect

to direct bandgap transition, for the NV center, we may imagine ZPL peak shift under the deep straining, opening new potential for highly efficient photonic device with reversible, modulated emission wavelength. Deep strain engineering is also a potential approach to turn the band structure and spin states of nitrogen-doped diamond. In addition to the predicted bandgap reduction, we also predicted symmetry breaking, band degeneracy and a reduction of total spin with applying deep compressive strain [100]. Since symmetry, energy degeneracy and spin transition are closely correlated, we would expect DESE to change the quantum properties of nitrogen doped diamond (and even NV center) substantially.

While diamond has been studied mostly as a bulk material, it can also be used in the form of membrane and bonded with various substrates for quantum applications [101]. Such deformation mode is similar with that of the two-dimensional material. Due to the thin thickness nature, large strain is more likely to be achieved and such a structure is suitable for flexible optoelectronic devices. This deformation process relies on the stable bonding between diamond and another different material as substrate [102, 103], and strains can be introduced via bending, stretching and pre-strain of the substrate material, such as the straining of diamond membrane with SnV center on fused silica [91], or diamond with SiV center on SiN [96], as shown in figures 3(c)–(e). In these two examples, strain plays an important role in increasing the ground state splitting (Δ_{GSS}), which is useful for improving the coherence time, especially for the high-temperature operation. Although only small strains (<1%) are introduced in the two examples, such prototype is expected to generate high static strain and enhance the spin coherence and improve the detection accuracy for more color centers of diamond.

5. Summary and outlook

With the development of diamond synthesis techniques and research on diamond nanomechanics and color centers, more and more quantum mechanical properties of diamond are being revealed and find applications in quantum sensing. Strain engineering is an emerging approach for tuning the quantum mechanical properties of diamond, but for most of the current studies, only small values of strain have been considered. Until now, deep strain has been achieved in diamond nano structures, laying solid foundation for deep strain induced physical properties modulation. Theoretical studies have predicted the great potential of deep strain engineering of diamond such as the bandgap reduction, indirect to direction bandgap transition, and symmetry-breaking induced spin degeneracy. We are expecting to see DESE induced wider ZPL shift and increase of spin coherence of more color centers of diamond, which should improve the detection accuracy, make more color centers useable and bring diamond to wider quantum sensing applications. Moreover, one may also expect the combination of strain and electrical fields, to tune the diamond quantum sensing performance together. Nevertheless, for the modification of quantum sensing properties of diamond by deep strain engineering, it is still a relatively new topic. In terms of the quantum application of deep strained diamond, there are still many challenges to be addressed such as how to generate and fix the high strains. We hope that more experimental and theoretical work will be conducted to shed more light on this topic in the near future.

Data availability statement

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

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