

Deep Spin Defects in Zinc Oxide for High-Fidelity Single-Shot Readout

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Wide-band gap oxides such as ZnO are favorable hosts for spin defect qubits due to their dilute nuclear spin background and potential for ultrahigh purity. Yet, a deep-level defect qubit with robust optical and spin properties has not been identified in this material. Here, using first-principles calculations, we predict that the molybdenum-vacancy complex, $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$, exhibits the essential characteristics of an optically addressable spin qubit: a spin-triplet ground state, visible-range optical transitions with high quantum

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yield, and an unusually small Huang-Rhys factor (~ 5 , compared to 10–30 in known ZnO defects). We further find long spin coherence times ($T_2 \sim 4$ ms) when both nuclear and impurity spin baths are considered, with paramagnetic impurities setting a threshold concentration of 0.035 ppm. Importantly, the combination of strong spin-orbit coupling and the absence of Jahn-Teller distortion supports spin-selective intersystem crossing and high-fidelity single-shot readout at elevated temperatures and across wide magnetic field ranges. By identifying ZnO as a host for deep-level defect qubits, our work points toward a pathway to scalable, integrable oxide-based quantum technologies and broadens the material foundation for solid-state quantum information science.

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I. INTRODUCTION

Point defects in wide-band gap solids with unpaired spins are emerging as promising candidates for quantum bits (qubits) due to their exceptional advantages such as elevated temperature operation and long coherence times. Consequently, the search for further spin qubit candidates has become a rapidly growing field, aiming to identify systems that can enhance existing quantum technologies or expand their capabilities. These efforts have led to the discovery of group-IV-vacancy qubits in diamond [1–4], vacancy spins in SiC [5,6], and more recently, boron vacancy spins in hexagonal boron nitride [7–9]. Notably, first-principles theoretical approaches have played a crucial role in this process by efficiently exploring the vast configuration space of defects in a material, narrowing down to the best candidates, and navigating the unknown connection between desirable qubit properties and the defect structure.

Finding optimal quantum defects for spin qubits requires evaluating both defect and host materials. An ideal host should have a wide band gap, which can span the energy range of ground and excited defect levels, minimal nuclear spin density for long spin coherence times, and can be grown as high-quality single crystals for minimum unwanted defects and impurities [10]. Given a host material, the best qubit candidates are identified based on deep defect levels, high spin states, strong radiative recombination rates compared to nonradiative ones, large Debye-Waller factor, and long spin relaxation and coherence times [11–13].

ZnO has recently gained recognition as an excellent host material due to its exceptional purity, achievable through molecular beam epitaxy (MBE) with sub-ppb background impurity concentrations [14–16]. This minimizes unintentional defects and paramagnetic noise, crucial for long coherence times. Furthermore, ZnO benefits from the inherent properties of oxides, where oxygen is 99.7% nuclear spin-free [17], ensuring a magnetically quiet environment and further reducing noise. The piezoelectricity property of ZnO introduces the possibility of strain tuning of the quantum states of spin defects.

Notably, several shallow donors in ZnO have already been identified as potential qubit candidates due to their

advantageous optical and spin properties. Neutral indium (In) and gallium (Ga) donor-bound electrons (D^0) form spin-1/2 qubit systems, exhibiting long spin relaxation times (T_1) (> 100 ms) [18,19], Hahn-spin-echo decoherence times ($T_2 = \sim 50$ μ s) [19], and narrow inhomogeneous linewidth [20,21]. Experimentally, all-optical methods have been utilized to control donor spin qubit states at cryogenic temperatures [19].

Expanding ZnO spin qubits beyond shallow donors is crucial for advancing ZnO-based spin-photon interfaces and exploring new capabilities. The low binding energy of shallow donors requires low-temperature operation [19,20,22]. Additionally, their UV-range optical emission is unsuitable for long-distance communications by optical fibers. Therefore, identifying deep-level defect qubits in ZnO is essential for room-temperature optical initialization and quantum emission in the IR and visible range that can be coherently coupled to the spin. Unlike spin-1/2 shallow donors, deep-level defects with a highly localized spin-triplet ground state would offer greater robustness against environmental noise, making them ideal for quantum sensing. Interestingly, several previous studies reported strong photon emission from deep-level defects in ZnO [23–27], but no spin qubits have been identified yet.

High-fidelity readout of spins is vital for spin-based quantum applications. Single-shot readout enables high-fidelity and efficient measurement, essential for quantum communication and algorithm execution [28–30]. A readout fidelity above 79%, corresponding to a signal-to-noise ratio greater than one, is widely accepted as the threshold for reliable single-shot detection [30]. This technique relies on resonant excitation of spin-selective, spin-conserving optical cycling transitions that are well isolated from intersystem crossings (ISCs), thereby enhancing both contrast and photon yield [31]. However, fidelity can be limited by spin leakage, where unwanted spin-flip transitions occur in the excited state due to magnetic noise or vibronic interactions. Previous studies have shown that reduced Jahn-Teller distortion, weak spin-phonon interactions, and large spin sublevel splittings help suppress spin leakage in systems such as SnV, SiV center in diamond, and divacancies in 4H-SiC [29,32–35].

In this article, we investigate nitrogen vacancy (NV) center like complex vacancy defects in ZnO and identify

the molybdenum vacancy complex $((\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+})$ as a promising spin qubit candidate with superior optical and spin properties. It features a highly polarized intersystem crossing with large nonaxial spin-orbit coupling (SOC) components and zero axial ones. Its strong spin-orbit-induced spin splitting in the excited state, along with a suppressed Jahn-Teller effect, allows minimal spin mixing even under magnetic fluctuations and elevated temperatures. Optically, it exhibits a remarkably low Huang-Rhys factor (~ 5), significantly lower than other known ZnO defects (typically > 10), resulting in a sharp zero-phonon line and high quantum yield due to suppressed electron-phonon coupling and phonon-assisted nonradiative processes. In addition, a long spin decoherence time (T_2) in ms order has been found, which could be limited by paramagnetic impurities even after isotopic purifications. These properties result in a promising qubit candidate for ultrahigh-fidelity spin readout.

II. RESULTS

In Fig. 1, we propose to design an NV^- -like C_{3v} defect complex in ZnO, consisting of an oxygen vacancy paired with a substitutional impurity (X) replacing a Zn atom [36]. To identify suitable spin qubit candidates, we follow the workflow outlined in Fig. 1, which begins with the search for defects exhibiting spin-triplet ($S = 1$) ground states and in-gap defect orbitals. Thermodynamic stability is then assessed via defect formation energies and charge transition levels.

Next, we assess the optical addressability of candidate defects using two main criteria: (a) radiatively allowed in-gap optical excitations characterized by the zero-phonon line (ZPL), absorption spectrum, and radiative lifetime; and (b) optimized electron-phonon coupling, evaluated via the Huang-Rhys (HR) factor, nonradiative

lifetime, quantum yield, and photoluminescence line-shape—ensuring efficient photon emission. We further assess spin decoherence arising from hyperfine interactions with nuclear spins and other paramagnetic defects, which ultimately determine the operational duration of the qubit for sensing and computational tasks.

Finally, we evaluate the electronic-spin multiplet structure and excited-state dynamics using a combination of multireference excited-state calculations, SOC, zero-field splitting (ZFS), and ISC rate predictions. Based on all these properties, we propose a promising optical readout protocol for spin defect qubits.

A. Defect Formation Energy

We computationally screen substitutional defects at Zn accompanied by an oxygen vacancy (V_{O}) across the periodic table, focusing on group B elements from groups III to VI and group A elements from groups I to V, guided by electronegativity and electron counting considerations [36]. For this purpose, we employ the hybrid density functional theory (DFT) at the Heyd-Scuseria-Ernzerhof (HSE) level of theory, which is robust for predicting ground-state properties. This leads to the identification of four triplet defect candidates: $(\text{X}_{\text{Zn}}\text{V}_{\text{O}})^q$, where q is the charge state (zero or positive for n-type defects), specifically $\text{Ti}_{\text{Zn}}\text{V}_{\text{O}}$, $(\text{V}_{\text{Zn}}\text{V}_{\text{O}})^+$, $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$, and $(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+$.

Figure 2(a) shows the defect charge transition levels (CTLs) for the four transition-metal-vacancy complex defects and intrinsic vacancies (V_{O} and V_{Zn}). The $2+$, $1+$, and $1+$ charge states, corresponding to the triplet ground state of $\text{Mo}_{\text{Zn}}\text{V}_{\text{O}}$, $\text{Nb}_{\text{Zn}}\text{V}_{\text{O}}$, and $\text{V}_{\text{Zn}}\text{V}_{\text{O}}$, respectively, are stable within the gap. These defects exhibit stable positive charge states at Fermi levels up to 0.2 eV below the conduction band minimum (CBM), primarily acting as electron donors.

The charge transition level $\varepsilon(2+/1+)$ for $\text{Nb}_{\text{Zn}}\text{V}_{\text{O}}$ and $\varepsilon(3+/1+)$ for $\text{V}_{\text{Zn}}\text{V}_{\text{O}}$ occurs at 0.76 and 1.63 eV below the CBM at 3.43 eV, respectively, while $\varepsilon(3+/2+)$ of $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ is 1.73 eV below the CBM, classifying them as deep donor defects in ZnO. In contrast, $\text{Ti}_{\text{Zn}}\text{V}_{\text{O}}$ has a $\varepsilon(1/0)$ transition at 0.85 eV above the CBM, which makes it unlikely to form in the neutral state. We therefore exclude $\text{Ti}_{\text{Zn}}\text{V}_{\text{O}}$ from further characterization.

As a benchmark, we also computed the CTL of the intrinsic defects. For V_{O} , we find the charge transition level $\varepsilon(2+/0)$ to be 2.26 eV above the valence band maximum (VBM). Our calculations also show an unstable $1+$ charge state, indicating a negative- U center, consistent with previously reported results using hybrid functionals [37–40]. For V_{Zn} , we observe that it is an amphoteric defect with stable charge states $2-$, $1-$, $1+$, and $2+$, in agreement with previous studies [38].

Figure 2(b) shows the formation energies of the aforementioned defects under O-poor and O-rich conditions.

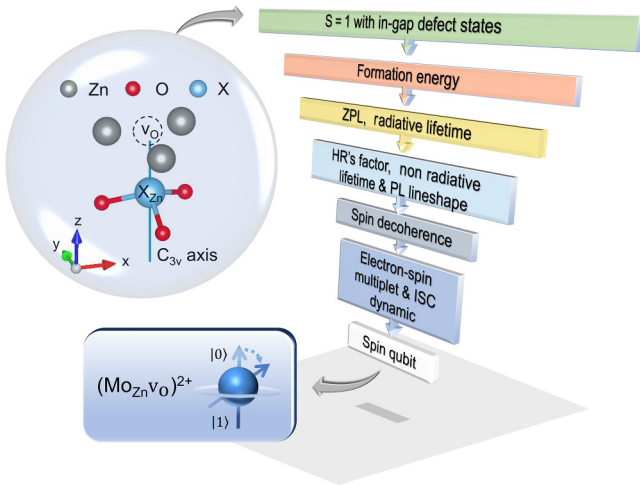


FIG. 1. The computational workflow of defect candidate search with critical parameters.

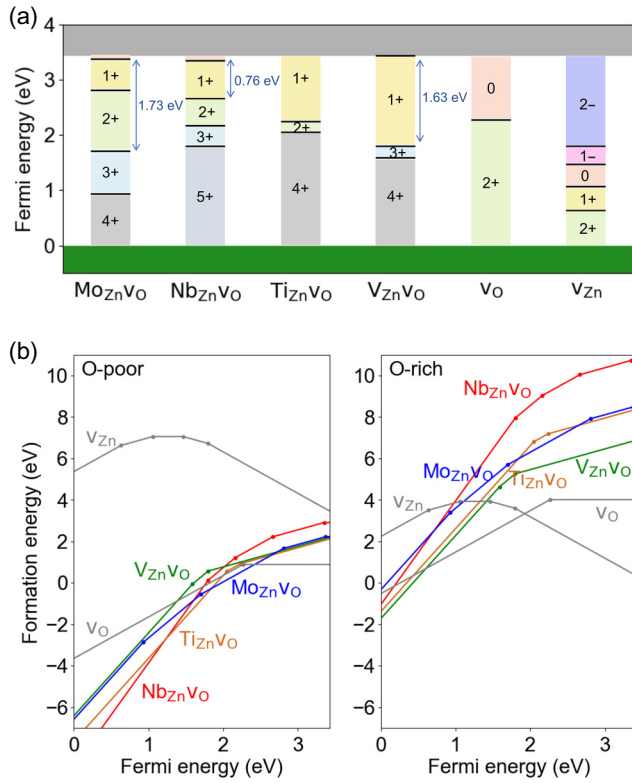


FIG. 2. Defect formation energies of complex vacancy defects. (a) Charge transition levels of the complex vacancy defects $X_{Zn}V_O$ ($X=Mo, Nb, Ti$, and V) and intrinsic defects V_O and V_{Zn} and (b) their formation energies at O-poor(left)/O-rich (right) conditions. The green/gray area in (a) corresponds to the valence/conduction band of ZnO, where VBM is set at 0 eV and the band gap is 3.43 eV.

The defect formation energies of all candidates are similar to the intrinsic donor V_O . In the n-type region (where the Fermi level close to CBM), the proposed defect formation energies are small under O-poor conditions and moderately large under O-rich conditions, compared to other intrinsic defects. In particular, intrinsic defects with smaller formation energies under O-rich conditions are mostly acceptors (V_{Zn}), which could compensate for the n-type donor defects. Additionally, we computed the formation energies of the isolated substitutional defect X_{Zn} , as shown in supplemental material, Fig. S1 [41].

We estimate the formation probabilities of $X_{Zn}V_O$ complexes in proportion to their isolated components $X_{Zn} + V_O$ based on the Boltzmann distribution at a representative synthesis temperature of 1200 K. In supplemental material, Fig. S2 [41], we show the complex vacancy formation probabilities in the oxygen vacancy-driven n-type Fermi-level range at 1200 K [42–44] (E_f between 2.26 and 3.43 eV above VBM, with CBM at 3.43 eV).

We find that the concentration of $Mo_{Zn}V_O$ increases from 0.97% with E_f at 2.26 eV to 3.96% with E_f at 3.43 eV. We note that at synthesis temperature, e.g., 1200 K, oxygen

vacancies can possibly overcome the ~ 1 eV activation barrier and diffuse to bind with substitutional defects X_{Zn} [43] to form $X_{Zn}V_O$.

Once the systems' temperature is lowered to the operating temperature (i.e., room temperature), the high activation barrier will lead to an extremely small kinetic rate and lock the defect complex to its thermodynamic equilibrium distribution at the synthesis temperature calculated above.

B. Single-particle defect levels

The single-particle defect levels and wave functions of the three candidates referenced to bulk ZnO band edges are shown in Figs. 3(a)–3(c), which provide insights of defect energy levels and molecular orbitals. To cross-check the results, we also performed the GW (G is Green's function and W is screened Coulomb interaction) [45] calculations, which describe more accurately quasi-particle band structure of solids, but are computationally much more demanding than the HSE calculations. We observed consistent results between HSE and GW calculations for the spin majority channel defect level, within 0.1 eV, as shown in Fig. 3(e), which support the use of HSE for this system's single-particle defect levels. More numerical details can be found in supplemental material, Sec. V [41].

All three candidates have similar defect-related orbitals, as shown in the schematic diagram of Fig. 3(d), which use the $(Mo_{Zn}V_O)^{2+}$ as an example. The a_1 and $2a_1$ states arise from the v_O -related states with a small mixing from the d_z orbital of substitutional transition metals. The doubly degenerate $e_{x,y}$ states come from the d_{xz} and d_{yz} orbitals of the transition metal, hybridized with the Zn dangling bond from v_O (more details for v_O and metal substitutional defects can be found in supplemental material, Sec. II [41]).

The $e_{x,y}$ orbitals in the spin-majority channel lie within the band gap of ZnO for $(Mo_{Zn}V_O)^{2+}$ and $(Nb_{Zn}V_O)^+$, while their counterparts in the spin-minority channel, and those for $(V_{Zn}V_O)^+$ in both channels, are more than 1.5 eV above the conduction band (CB). This results from Hund's rules, where the d orbital fillings favor open-shell configurations, causing the occupied $e_{x,y}$ orbitals in the spin-majority channel to have lower in-gap energy levels. We note that the oxidation state of the transition metal in the complex defect differs from the overall charge state q of the complex. Based on electron counting and orbital analysis, the oxidation state of the transition metal ions is identified as Mo^{+4} , Nb^{+3} , and V^{+3} in their respective complex structures, all corresponding to a d^2 electron configuration.

C. ZPL, absorption spectra, and radiative lifetime

In this section, we begin our investigation of the spin-photon interface by determining the dipole moments, zero-phonon line (ZPL) energies, and radiative lifetimes (τ_R) of the defect candidates. These quantities provide the key

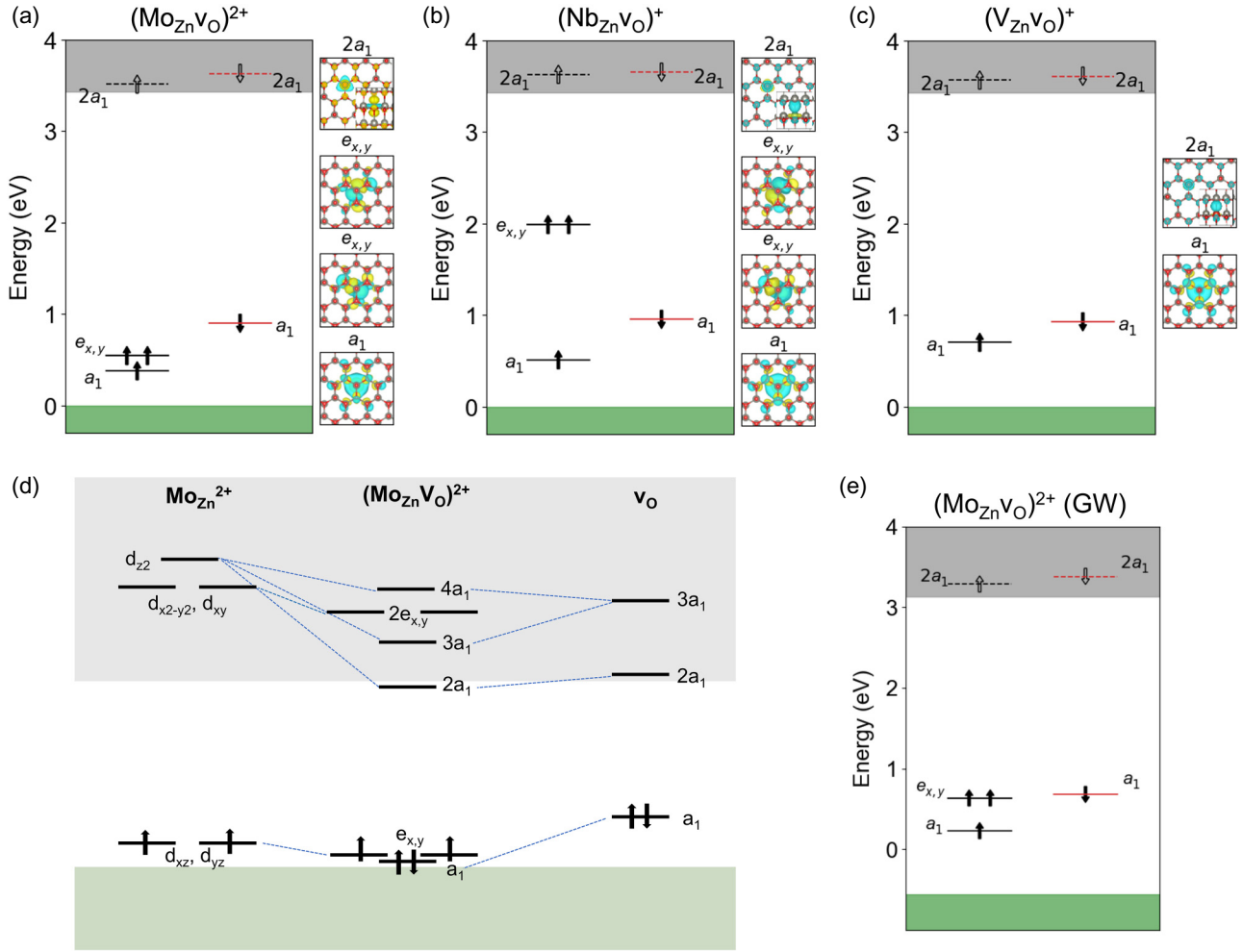


FIG. 3. Single-particle levels and wavefunctions. Single-particle defect levels [horizontal black (majority spin) and red lines (minority spin)] of the (a) $(\text{Mo}_{\text{Zn}}\text{V}_\text{O})^{2+}$, (b) $(\text{Nb}_{\text{Zn}}\text{V}_\text{O})^+$, (c) $(\text{V}_{\text{Zn}}\text{V}_\text{O})^+$, and (e) v_O defects in ZnO, calculated at the Heyd-Scuseria-Ernzerhof (HSE) ($\alpha = 0.375$). The green/gray area corresponds to the valence/conduction band of ZnO, respectively. The orbitals labeled with the same symmetry symbol across three defects share the same feature and wave function natures. (d) is the schematic diagram showing the origin of defect states of $(\text{Mo}_{\text{Zn}}\text{V}_\text{O})^{2+}$ from v_O and $(\text{Mo}_{\text{Zn}})^{2+}$, respectively. (e) Quasiparticle bandstructure at G_0W_0 [45] with the PBE0 starting point for $(\text{Mo}_{\text{Zn}}\text{V}_\text{O})^{2+}$.

inputs for estimating the brightness and spin-dependent optical contrast of the defect, which in turn determine the achievable fidelity of optical initialization and readout.

First, we compute the absorption spectra of the triplet states within the Random-Phase Approximation (RPA) using single particle wavefunctions computed at HSE (Fig. S5 in the supplemental material [41]). Local-field effects [46] were included, as they are crucial in nonhomogeneous dielectric environments such as defect systems. The defect transition energies are well separated from the bulk excitation, spanning from ultra-red to blue. The squared dipole moments (μ^2) for defect transitions, for light polarized along the x , y , and z directions, were extracted from the spectra. These values are summarized in Table I, where the relevant polarization direction for each non-negligible μ^2 is indicated by “ $x/y/z$ ” in the transition

column. We note that these computed dipole moments are consistent with the group-theory analysis for C_{3v} , where the $e \rightarrow 2a_1$ transition is allowed only when light is polarized in the x - y plane for the majority spin channel, while the $a_1 \rightarrow 2a_1$ transition is allowed only when light is polarized along the z direction for the minority spin channel.

We next evaluate the zero-phonon line (ZPL) energies, defined as the energy difference between the minima of the excited- and ground-state potential surfaces. For accurate prediction and cross-validation, we employed three different computational methods: (i) multideterminant constrained DFT (mcDFT; details in Sec. VD), (ii) quantum defect embedding theory (QDET) [47–50], and (iii) the multiconfigurational self-consistent field method with perturbative correction (DLPNO-NEVPT2@CASSCF) [51]

TABLE I. Summary of the optical properties of the proposed defects in their triplet states. The zero-phonon line values listed represent the finalized choices obtained from the selected levels of theory, with DLPNO-NEVPT2@CASSCF used for the spin-majority transition of $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ and multideterminant constrained density functional theory (mcDFT) used for all remaining transitions. Also reported are the relaxation energy computed with multideterminant constrained density functional theory (E_{rel}), the squared transition dipole moment (μ^2), and the radiative lifetime (τ_R) for each ZnO defect system.

Spin	Transition	ZPL (eV)	E_{rel} (eV)	μ^2 (bohr ²)	τ_R (μs)
$(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$					
Maj	$e \rightarrow a_1$ (x,y)	2.08	0.18	4.96×10^{-2}	5.03
Min	$a_1 \rightarrow 2a_1$ (z)	1.56	0.85	2.20×10^{-2}	27.10
$(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+$					
Maj	$e \rightarrow a_1$ (x,y)	1.07	0.51	8.09×10^{-2}	36.8
Min	$a_1 \rightarrow 2a_1$ (z)	1.72	0.74	2.82×10^{-2}	15.7
$(\text{V}_{\text{Zn}}\text{V}_{\text{O}})^+$					
Maj	$a_1 \rightarrow 2a_1$ (z)	0.13	2.49	6.09×10^{-2}	1.69×10^4
Min	$a_1 \rightarrow 2a_1$ (z)	1.62	0.86	2.19×10^{-3}	244.00

(details in supplemental material, Secs. V and XI [41]). The mcDFT method, while mean-field in nature, provides a practical balance between accuracy and computational cost, whereas QDET and DLPNO-NEVPT2@CASSCF are higher-level approaches better suited for strongly correlated open-shell excited states of transition-metal defects in oxides.

Table II summarizes the energies of the triplet ground state 3A_2 , the triplet excited state (3E), and the singlets (1E and 1A_1) for $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$, where the doubly degenerated many-body 3E state is characterized by the $e \rightarrow a_1$ transition, and the detail of single particle configuration for states can be found in Sec. VD and Table S11 [41]. The three approaches yield consistent results for the 3E triplet excited state and the 1E singlet, confirming that mcDFT can reliably capture the excited-state properties

TABLE II. Computed zero-phonon line energies (eV) of $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ in ZnO at different levels of theory.

Theory	ZPL energies (eV) ^a			
	3A_2	1E	1A_1	3E
mcDFT@HSE	0	0.83 ^b	—	1.99 ^c
QDET(14e14o)	0	—	—	1.88
DLPNO-NEVPT2@CASSCF(4,7)	0	0.81	1.63 ^d	2.08

^aFor each theory, the ZPL energy is calculated as the vertical excitation energy (with a theory stated on the left column) at the fixed ground-state geometry minus the excited-state relaxation energy calculated from mcDFT.

^bThe relaxation energy of 1E from mcDFT is 0.12 eV.

^cThe relaxation energy of 3E from mcDFT is 0.18 eV.

^dThe geometry of 1A_1 is assumed to be the same as 3A_2 due to the similar nondegenerate symmetry.

of this system. A more detailed comparison of the optical properties across methods is provided in supplemental material, Sec. XIV [41]. For the remainder of our study, we adopt the DLPNO-NEVPT2@CASSCF result as the ZPL reference for our key defect candidate, $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$, while using mcDFT values for other defects as a balance between accuracy and computational efficiency. The final ZPL results are summarized in Table I, and these values are used throughout the remainder of this work whenever a ZPL input is required.

Based on the ZPL energy and transition dipole moment, we calculate radiative lifetimes (τ_R) scales as $E^{-3}\mu^{-2}$ [Eq. (4)] for $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+$, as summarized in Table I (details in supplemental material, Sec. IV [41]). For example, the $e \rightarrow 2a_1$ transition in $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ has the shortest lifetime ($\sim 5.03 \mu\text{s}$). Comparing with other spin defect qubits, the radiative lifetimes of the proposed defects are longer than the NV center (12 ns) [52], but shorter than the boron vacancy in hBN (20 μs) [53]. The difference is mainly due to variations in dipole moments based on Eq. (4), i.e., the NV center has a transition dipole moment ($\mu^2 \sim 3.87 \text{ a.u.}$ [54]), much larger than that of our proposed systems ($\mu^2 \sim 10^{-2} \text{ a.u.}$), while the boron vacancy exhibits a much smaller one ($\mu^2 \sim 10^{-4} \text{ a.u.}$ [55]).

D. Huang-Rhys factor, nonradiative recombination, and photoluminescence lineshape

To further characterize the phonon-related optical properties of the defect candidates, we compute the Huang-Rhys (HR) factors [56,57], the nonradiative lifetime (τ_{NR}) [58], as well as the quantum yield, as summarized in Table III. The HR factor quantifies the strength of electron-phonon coupling and the energy reorganization during electronic transitions. Small electron-phonon coupling

TABLE III. The phonon-related properties of optical spectroscopy for $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+$. ΔQ is the mass-weighted displacements between the excited and ground-state geometries. $k \approx \text{ZPL}/\hbar\omega_{\text{eff}}(\text{gs})$ is used to approximate the number of phonons necessary to mediate the transition. S_f^{eff} is the HR's factor computed by effective phonons for ground state. τ_{NR} is the nonradiative lifetime. W_{if} and X_{if} are the electronic part and phonon part of the nonradiative lifetime (τ_{NR}). QY is the quantum yield.

Spin	Transition	ZPL (eV)	ΔQ (amu ^{1/2} Å)	k	τ_{NR} (μs)	W_{if} (eV/(amu ^{1/2} Å))	X_{if}	S_f^{eff}	QY
$(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$									
Maj	$e \rightarrow 1a_1$ (x,y)	2.08	1.19	72	Not allowed	5.13×10^{-2}	1.24×10^{-29}	4.96	High (≈ 1)
Min	$1a_1 \rightarrow 2a_1$ (z)	1.56	5.05	91	1.07×10^{-7}	3.75×10^{-2}	1.23×10^{-1}	50.64	Small (≈ 0)
$(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+$									
Maj	$e \rightarrow 1a_1$ (x,y)	1.07	1.80	31	1.63×10^{-9}	4.42×10^{-2}	5.82	15.36	Small (≈ 0)
Min	$1a_1 \rightarrow 2a_1$ (z)	1.72	3.83	84	2.47×10^{-5}	4.62×10^{-2}	3.52×10^{-4}	35.67	Small (≈ 0)

strength gives a small HR factor, which leads to the relative high intensity of the ZPL compared to the phonon sideband (PSB). Nonradiative (NR) recombination refers to the electron-hole recombination mediated by phonons, which does not contribute to photoluminescence. The optical brightness, determined by the competition between the radiative and NR recombination paths, is characterized by the quantum yield ($\text{QY} = (1/\tau_{\text{R}})/(1/\tau_{\text{R}} + 1/\tau_{\text{NR}})$).

Among the spin defects we studied in Fig. 3, all the $a_1 \rightarrow 2a_1$ transitions show large HR factors and fast non-radiative decay, leading to a large PSB and low quantum yield. This is consistent with the excited-state relaxation energy (E_{rel}) prediction in Table I, where the E_{rel} for the $a_1 \rightarrow 2a_1$ transition are generally larger than those for the $e \rightarrow a_1$ transition, indicating stronger electron-phonon coupling. From the orbital perspective, the $a_1 \rightarrow 2a_1$ transition in all three systems is mainly originated from the internal transition of V_{O} defect, with small mixing from the d_z orbital of the transition metal (TM) ion, as shown in Fig. 3(d), which exhibits significant electron-phonon coupling and a large lattice relaxation energy (~ 1 eV). We also note that this large electron-phonon coupling associated with the Zn dangling bonds is consistent with previous study [59]. Based on these considerations, we exclude all spin-minority transitions as well as the $(\text{V}_{\text{Zn}}\text{V}_{\text{O}})^+$ defect from the list of viable candidates.

The spin-majority transitions of $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+$ have smaller HR factors (4.96 and 15.36, respectively) compared to the spin-minority channels. However, $(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+$ still shows a fast nonradiative decay and low quantum yield. In contrast, $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ combines a small HR factor with slow nonradiative decay, making it the best candidate.

To gain insights on determining factors of HR factors, we compare the two proposed defects with others in ZnO and various host materials in Table IV. We observe that host materials with light elements (C, BN) generally exhibit smaller HR factors compared to ZnO or GaN, which can be attributed to the dependence of atomic mass (m_{α}) in ΔQ , hence HR factor S_f^{eff} ($S_f^{\text{eff}} = \omega_{\text{eff}}\Delta Q^2/2\hbar$

and $\Delta Q = \sqrt{\sum_{\alpha,t} m_{\alpha} \Delta R_{\alpha,t}^2}$ [56]). Furthermore, the dominant phonon frequency (ω_{eff}) varies within a factor of 2, particularly for defects in ZnO. Thus, the primary factor influencing HR variation within a given host (e.g., ZnO) is the displacement between excited and ground states ($\Delta R_{\alpha,t}$).

Our proposed $(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}$ defect (spin-majority channel $e \rightarrow 1a_1$ transition) exhibits a surprisingly small HR factor of 4.96, which, while slightly larger than that of the NV center in diamond, remains one of the smallest among known defects in ZnO. In comparison, experiments have successfully resolved the zero-phonon line (ZPL) of copper substitutional defects in ZnO, which have an HR factor

TABLE IV. A summary of HR factor (S) of various defects from past literature and this work.

Defect(host)	$\Delta Q(\text{amu}^{1/2} \text{Å})$	$\hbar\omega(\text{meV})$	S
$(\text{Mo}_{\text{Zn}}\text{V}_{\text{O}})^{2+}(\text{ZnO})$	1.19	29	4.96
$(\text{Nb}_{\text{Zn}}\text{V}_{\text{O}})^+(\text{ZnO})$	1.80	40	15.36
Li_{Zn} [60](ZnO)	3.22	36	28
N_{O} [61](ZnO)	1.92	40	15.3
$\text{Cu}_{\text{Zn}}^{+ \rightarrow 0}$ [62](ZnO)	$\approx 1.39^a$	57	13.1
$\text{Cu}_{\text{Zn}}^{+ \rightarrow 0}$ [62](ZnO)	$\approx 1.33^a$	52	11.0
$\text{V}_{\text{Zn}}\text{Al}_{\text{Zn}}^{0 \rightarrow -}$ [63](ZnO)	2.7	33	25
$\text{V}_{\text{Zn}}\text{Si}_{\text{Zn}}^{+ \rightarrow 0}$ [63](ZnO)	2.7	33	25
$\text{V}_{\text{Zn}}\text{H}^{0 \rightarrow -}$ [63](ZnO)	2.95	30	27
C_{N} [60](GaN)	1.61	42	10
$\text{Zn}_{\text{Ga}}\text{V}_{\text{N}}$ [60](GaN)	3.33	26	30
NV^- [64](Diamond)	$\approx 0.7^a$	64	3.67
Cl-V [65](4H-SiC)	$\approx 0.96^a$	32 to 40	3.5 to 4.4
VB^- [53,55](hBN)	$\approx 1.1^a$	24.8	3.69
$\text{C}_{\text{B}}\text{C}_{\text{N}}$ [66](hBN)	$\approx 0.38^a$	120	2
$\text{N}_{\text{B}}\text{V}_{\text{N}}$ [67](hBN)	0.66	30, 46, 48 ^b	4.49

^a ΔQ is estimated by S, through $S = \omega_{\text{eff}}\Delta Q^2/2\hbar$.

^bphonon modes which have the largest contribution to the HR factor; the rest of $\hbar\omega$ in the table are effective phonon frequencies.

of 11 [62]. This suggests that the ZPL of $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ should also be resolvable experimentally.

To understand why $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ defect has a smaller HR factor than other defects in ZnO, we compare $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ with $(\text{Nb}_{\text{Zn}}\text{VO})^+$ as an example. In Figs. 4(d) and 4(e), we show the effective phonon mode and the contribution to ΔQ^2 from atoms in real space for the spin majority transition of $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{VO})^+$. Both of the two systems have an effective phonon mode localized within 5 Å from the TM defect center. The major contributors to ΔQ^2 arise from the Zn atoms around the vacancy and next-nearest neighbors of the TM substitution site. With similar atomic mass contributions (single atom Mo or Nb mass difference is negligible in the summation in ΔQ), the smaller ΔQ^2 of $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ is mostly from overall smaller atomic displacement (ΔR_α) between ground and excited states. This difference between two defects may arise from the closer ionic radius between Mo (oxidation state +4, coordination VI, 0.65 Å) and Zn (oxidation state +2, coordination IV, 0.60 Å), compared to Nb (oxidation state +3, coordination VI, 0.72 Å) [68].

We next discuss photoluminescence (PL) lineshape and phonon sidebands of the $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{VO})^+$ spin-majority transitions using the generating function approach

with all phonons [61]. As shown in Figs. 4(c) and 4(f), the $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ exhibits a sharp and high intensity ZPL peak and a phonon side band with an anti-Stokes (AS) shift of 0.127 eV. On the other hand, the ZPL of $(\text{Nb}_{\text{Zn}}\text{VO})^+$ is relatively small, but its AS shift of 0.348 eV may allow it to be distinguishable from the large PSB. The corresponding partial HR factor spectrum functions are shown in Figs. 4(d) and 4(j). We found that $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ has a dominant peak at 21 meV, and $(\text{Nb}_{\text{Zn}}\text{VO})^+$ between 19–30 and 55–65 meV. To understand the localization of these phonon modes, we compute the inverse participation ratio (IPR), estimating the number of atoms involved in the vibration of the phonon modes [69]. We found that the IPRs are all larger than 20 for the two defects, representing delocalized bulk-like phonon modes. Their energies lie within the acoustic band of bulk ZnO.

In the Franck-Condon picture, the AS shift corresponds to the ground-state energy difference between its equilibrium geometry and excited-state geometry. We found their values (0.13 eV for $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and 0.35 eV for $(\text{Nb}_{\text{Zn}}\text{VO})^+$) to be close to the values from CDFT relaxation energy of ground state, i.e., 0.14 for $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and 0.50 eV for $(\text{Nb}_{\text{Zn}}\text{VO})^+$, respectively. In addition, the HR factor obtained by all-phonon-mode summation is

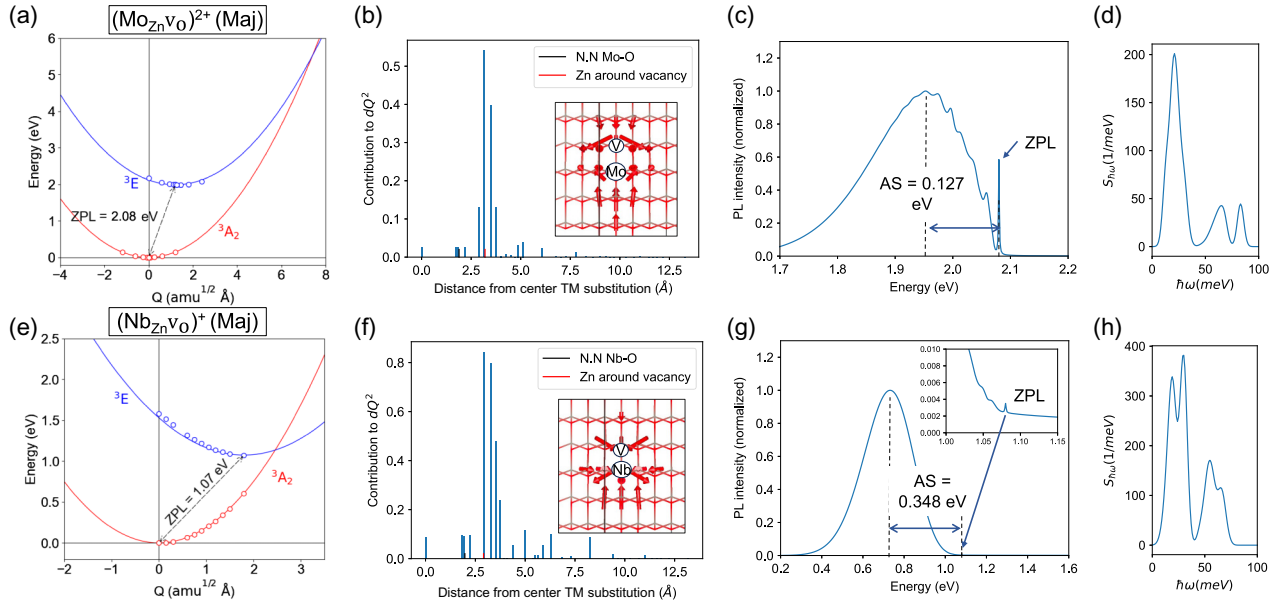


FIG. 4. Electron-phonon properties of $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{VO})^+$ spin majority transition between triplet states. The panels (a)–(d) are for $e \rightarrow a_1$ transition in $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and (d)–(g) are for $e \rightarrow a_1$ transition in $(\text{Nb}_{\text{Zn}}\text{VO})^+$. (a) and (d) are the configuration coordinate diagram of the potential surface (dots) fitted by harmonic potentials with effective phonon modes (lines). (b) and (e) are the module square of mass weighted displacement dQ^2 distribution for proposed defects contributed from atoms as a function of radius from the TM defect center. The contribution to dQ^2 from atom at position r from the TM site is defined as $dQ^2(r) = \sum_{\alpha} dQ^2(\alpha)|_{|r_{\alpha}-r|<0.1}$, where the r_{α} is the atom α 's distance from TM site and 0.1 is the step size of the sampling. The total displacement $\Delta Q = \int dQ^2(|\vec{r}|) d\vec{r}$. (c) and (f) are the photoluminescence lineshape, where the anti-Stokes shifts are determined by the energy difference between the highest peak of phonon side band and the zero-phonon line. (d) and (j) are the corresponding phonon spectrum functions revealing the partial HR factor contributed by the phonon mode with energy $\hbar\omega$.

in reasonable agreement with the one with an effective phonon along the configuration coordinate [58], which validates the consistency between different theoretical approaches in this system (more details can be found in Sec. VIII of the supplemental material [41]).

Finally, to explain why the spin-majority nonradiative (NR) transition is forbidden in $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ but fast in $(\text{Nb}_{\text{Zn}}\text{VO})^+$, we refer to classical Marcus theory in the high-temperature limit, as illustrated in Figs. 4(a) and 4(d). In this picture, the potential energy surface (PES) crossing between the ground state (red curve) and the excited state (blue curve) defines an energy barrier that the phonon-assisted NR recombination process needs to overcome. A much larger barrier is obtained for $e \rightarrow 2a_1$ transition in $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ than $(\text{Nb}_{\text{Zn}}\text{VO})^+$, which explains the much slower NR process in $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$. This could be understood from the much larger ZPL and smaller ΔQ in $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$.

From a theoretical perspective, the phonon-assisted NR recombination rate, within the static coupling approximation and the one-dimensional effective phonon approximation, is expressed with the product of two terms: the square of electronic term (W_{if}) and the phonon term (X_{if}) (more details in Sec. V E) [58]. The electronic coupling term exhibits relatively small variation between two defects as shown in Table III. The phonon term X_{if} of $(\text{Nb}_{\text{Zn}}\text{VO})^+$ is many orders of magnitude larger than that of $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$. Consequently, $(\text{Nb}_{\text{Zn}}\text{VO})^+$ exhibits stronger electron-phonon coupling and a faster NR decay. More detailed discussion can be found in supplemental material, Fig. S9 [41].

E. Spin decoherence time

In order to predict spin decoherence time of qubit candidates, we compute the free-induction decay (FID) signals due to hyperfine couplings with the nearby nuclear spins of ^{67}Zn ($I=5/2$, 4.1%) and ^{17}O ($I=5/2$, 0.038%). Figures 5(a) and 5(b) show the FID signals for $(\text{Nb}_{\text{Zn}}\text{VO})^+$ and $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$, respectively, under three different external magnetic field strengths. We observe that the FID signals saturate at 300 G, with maximum decay time (T_2^*) of 0.40 and 0.37 μs for $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{VO})^+$, respectively. Both exhibit strong non-Gaussian features with stretched exponents of 0.51 and 0.56 for $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ and $(\text{Nb}_{\text{Zn}}\text{VO})^+$, respectively, due to a strong anisotropic hyperfine coupling [70]. We remark that the T_2^* times of these qubit candidates are an order of magnitude smaller than that of diamond NV centers [71] and of a silicon vacancy in SiC [72] due to the larger magnetic moment and abundance of ^{67}Zn nuclear spins than those of ^{13}C nuclear spins (1.1%) in diamond. However the T_2^* is order of magnitude larger than the shallow donor in ZnO [19].

The inhomogeneous broadening effect in FID can be removed by applying the Hahn-echo pulse sequence.

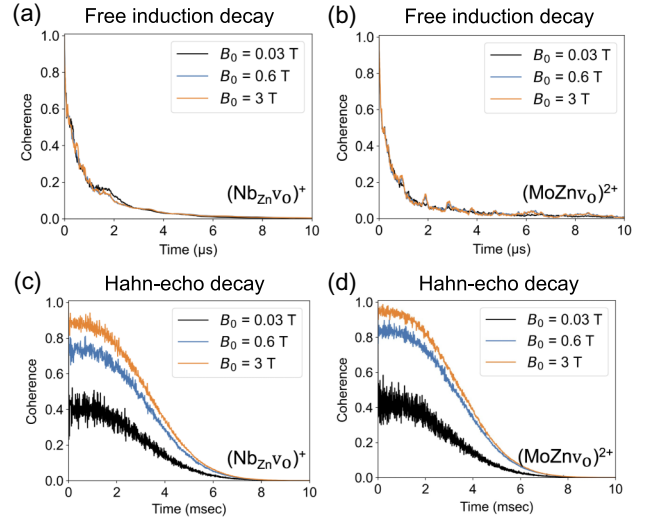


FIG. 5. Spin decoherence of transition metal-vacancy complex in a nuclear spin bath ZnO. (a) and (b) Free induction decay (T_2^*) of $(\text{Nb}_{\text{Zn}}\text{VO})^+$ (a) and $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ (b) in ZnO under various external magnetic field strengths (B_0). (c) and (d) Hahn-echo coherence decay (T_2) of $(\text{Nb}_{\text{Zn}}\text{VO})^+$ (c) and $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$ (d) in ZnO under various external magnetic field strengths.

Figures 5(c) and 5(d) show the computed Hahn-echo signals of the $(\text{Nb}_{\text{Zn}}\text{VO})^+$ and $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$, respectively, under three different magnetic field strengths. Notably, we observe that the decoherence occurs on two different time scales: an initial partial collapse within the μs range, followed by a gradual decay on the millisecond timescale. The gradual coherence decay is due to the dynamical fluctuation of the nuclear spin bath arising from the magnetic dipolar coupling between nuclear spins [70]. We find that the gradual decay saturates above 300 G and is described by a stretched exponential function with exponents of 2.49 (2.32) and the T_2 times of 4.04 (3.96) ms for $(\text{Mo}_{\text{Zn}}\text{VO})^{2+}$, and $(\text{Nb}_{\text{Zn}}\text{VO})^+$ in the parentheses, respectively. This saturation is attributed to the suppression of spin-flip transitions other than the spin flip-flop transitions in the bath, caused by large energy gaps between nuclear spin levels due to the Zeeman effect [70]. For the early partial collapse observed in Figs. 5(c) and 5(d), we attribute its origin to strain-induced inhomogeneous quadrupole interactions around the defect site (see supplemental material [41], Table S9). The variation in quadrupole interactions leads to differing precession frequencies of the nuclear spins [73], causing irregular electron spin echo envelope modulation (ESEEM) [74–76]. In supplemental material [41], Fig. S12, we demonstrate that this early coherence collapse disappears in a hypothetical bath model, where quadrupole interactions are removed from the spin Hamiltonian, confirming that the collapse is due to quadrupole interactions. Additionally, the partial coherence collapse disappears above 3 T, where the Larmor frequency becomes

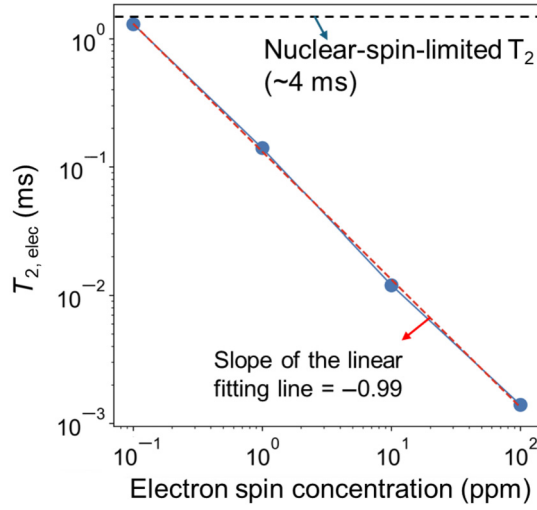


FIG. 6. Electronic spin-induced decoherence time T_2 as a function of electron spin concentrations. The black-dashed line shows the T_2 time determined by the nuclear spin bath in ZnO and the red-dashed line represents a linear fitting model in logarithmic scale.

the dominant frequency of the nuclear spins, suppressing the ESEEM depth.

In addition to nuclear spins, ZnO contains intrinsic defects, some of which may be paramagnetic [77–80], even in isotopically purified materials. In such engineered samples, where nuclear spins are removed, qubit decoherence would be dominated by the electron spin bath from paramagnetic defects. To quantify this effect, we compute the Hahn-echo T_2 time as a function of electron spin-1/2 concentration in ZnO, ranging from 0.1 to 100 ppm ($10^{16} - 10^{19} \text{ cm}^{-3}$), which is consistent with experimental defect concentrations [81,82]. As shown in Fig. 6, when the electron spin concentration reaches 0.035 ppm ($2.85 \times 10^{15} \text{ cm}^{-3}$), the T_2 time induced by the electron spin bath becomes comparable to that caused by the nuclear spin bath. Our results suggest that for isotopic purification to be effective, the paramagnetic defect concentration should be kept below 0.035 ppm.

As the electronic spin concentration increases beyond 10^{16} cm^{-3} , the T_2 time is rapidly reduced to sub-millisecond. We find that the T_2 time decreases almost linearly in the log scale with a slope of -0.99 . We note that nuclear spins can be removed or significant reduced by nuclear spin purification, then the paramagnetic impurities would be the main source for spin decoherence.

F. Spin properties and optical read-out

The comprehensive analysis of the thermodynamic properties, optical properties, and electron-phonon coupling, as well as spin decoherence time of these transition

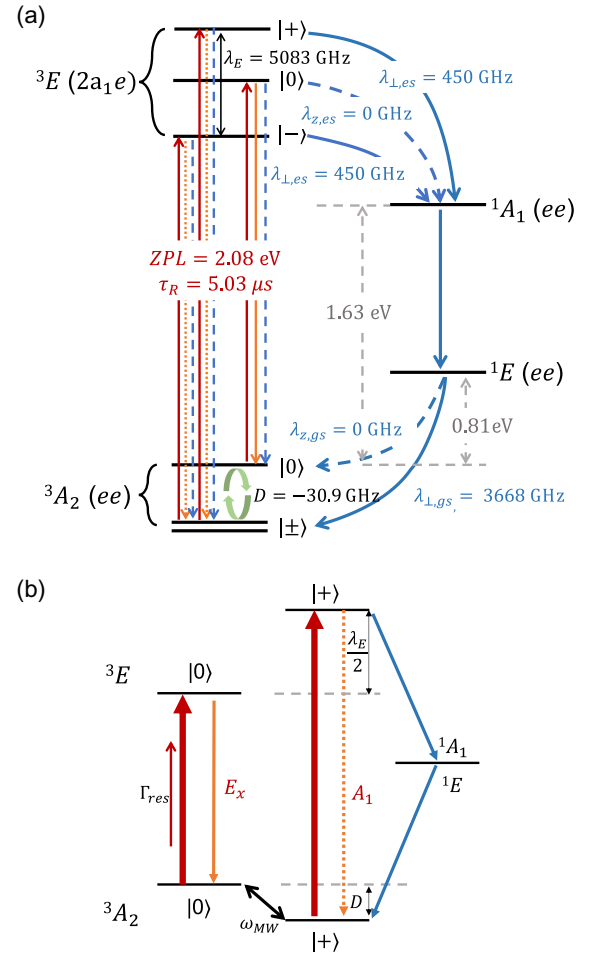


FIG. 7. Spin properties and optical read-out of $(\text{MoZnVO})^{2+}$ in ZnO. (a) Energy diagram and excited-state dynamics of $(\text{MoZnVO})^{2+}$ in ZnO. (b) A schematic diagram for single shot readout with qubit encoded in $|0\rangle$ and $|+\rangle$. (For qubit encoded in $|0\rangle$ and $|-\rangle$, the only difference is that the $\lambda_E/2$ become negative.) Γ_{res} denotes resonant excitation of the $|0\rangle$ state or the $|\pm 1\rangle$ state. In both panels (a) and (b), the red lines represent the optical excitation, the blue lines represent the phonon-assisted nonradiative recombination processes, and the orange lines represent the radiative recombination processes. Solid lines indicate dominant processes, dashed lines indicate forbidden ones, and dotted lines indicate allowed but less dominant processes. In panel (a), we show all these processes, while in panel (b), we keep only the allowed ones.

metal vacancy complexes in ZnO demonstrated their suitability for optically addressable spin qubits. $(\text{MoZnVO})^{2+}$ stands out to be the most promising, with its low formation energy and optimal combination of other characteristics. We therefore focus on discussing the readout of spin qubit $(\text{MoZnVO})^{2+}$ through a spin-photon interface as follows.

Figure 7(a) provides the multiplet structure diagram of $(\text{MoZnVO})^{2+}$ in ZnO that consists of a triplet ground state (3A_2), triplet excited state (3E), and two singlet shelving

TABLE V. Calculated zero-field splitting (ZFS) and spin-orbit coupling (SOC) parameters of $(\text{MoZnVO})^{2+}$ and NV center in diamond using SA(6)-CASSCF(6,6) [54]. All values are in GHz. The λ are the SOC parameters between states, where $\lambda_E = \langle {}^3E^\pm | H_{\text{SOC}} | {}^3E^\mp \rangle$, $\lambda_{z,gs} = \langle {}^1E | H_{\text{SOC}} | {}^3A_2^0 \rangle$, and $\lambda_{\perp,gs} = \langle {}^1E | H_{\text{SOC}} | {}^3A_2^\pm \rangle$, $\lambda_{z,es} = \langle {}^3E^0 | H_{\text{SOC}} | {}^1A_1 \rangle$, $\lambda_{\perp,es} = \langle {}^3E^\pm | H_{\text{SOC}} | {}^1A_1 \rangle$. The superscript on the right side of each state denotes the spin sublevel m_s .

	D_{SS}	D_{SOC}	D	λ_E	$\lambda_{z,gs}$	$\lambda_{\perp,gs}$	$\lambda_{z,es}$	$\lambda_{\perp,es}$
$(\text{MoZnVO})^{2+}$	4.70	-35.60	-30.90	5082.68	0.02	3668.05	0.02	450.48
NV ⁻ [54,84]	3.03	0.04	3.08	14.21	0.03	5.22	0.06	3.96

states (1E and 1A_1), a four-level structure similar to NV⁻ in diamond [83].

The degeneracy of the ground-state spin triplet manifold is split by intrinsic spin-spin and spin-orbit interactions. This splitting is characterized by the axial zero-field splitting (ZFS) parameter D (between $|m_s = 0\rangle$ and $|m_s = \pm 1\rangle$) and rhombic E (between $|m_s = +1\rangle$ and $|m_s = -1\rangle$). E is zero at C_{3v} symmetry. We obtain the ground-state ZFS by perturbation theory with first-order spin-spin contribution [84,85] $D_{SS} = 4.7$ GHz and second-order spin-orbit contribution [86] $D_{\text{SOC}} = -35.6$ GHz, respectively. In comparison, we obtain the $D_{\text{SOC}} + D_{SS} = 21.00$ GHz for $(\text{VZnVO})^+$, in reasonable agreement with the experimental value of 22.37 GHz [87,88] for V^{3+} in ZnO (see Sec. X in the supplemental material [41]).

The SOC parameters can be used to determine the fine structure of the spin state and the spin selectivity of optical transitions [89]. At the C_{3v} symmetry, the spin-orbit coupling Hamiltonian can be written in terms of its axial (λ_z) and nonaxial ($\lambda_\perp$) components,

$$H_{\text{soc}} = \frac{\lambda_\perp}{2} (L_+ S_- + L_- S_+) + \lambda_z L_z S_z, \quad (1)$$

where $L_\pm, L_z$ and $S_\pm, S_z$ are the ladder operators of orbital and spin angular momentum, respectively, in the two-electron spin system with $|S, m\rangle$ basis. The axial component λ_z can split the degenerate sublevels within the excited-state triplet manifold [83]. It is also associated with the transitions between the triplet $m = 0$ spin sublevels and the singlet state. The nonaxial component $\lambda_\perp$ is associated with the transitions between the triplet $m = \pm 1$ spin sublevels and the singlet state.

Given the multireference nature of excited states of $(\text{MoZnVO})^{2+}$, we calculate the SOC matrix elements with the CASSCF method, which was shown to provide accurate SOC for multireference states [54]. The computed ZFS and SOC parameters are shown in Table V. In contrast to the ground state, where the first-order SOC contribution vanishes, the spin splitting of the 3E excited state is dominated by a strong first-order SOC interaction, with $\lambda_E = \langle {}^3E^\pm | H_{\text{SOC}} | {}^3E^\mp \rangle = 5083$ GHz, consistent with the group theory prediction [83]. We noted that the ratios $\lambda_E({}^3E)/D_{\text{SOC}}({}^3A_2)$ for both the NV center and $(\text{MoZnVO})^{2+}$ are on the order of 10^2 [54] in Table V.

We find large nonaxial component for both excited-state and ground-state intersystem crossings as shown in Table V and Fig. 7. This large SOC originates from the transition metal ion Mo^{4+} as a part of the vacancy defect. The corresponding SOC parameters in NV center are two orders smaller than $(\text{MoZnVO})^{2+}$. This indicates that the ISC in $(\text{MoZnVO})^{2+}$ could be faster than the NV center in diamond, as the large SOC likely leads to a fast ISC [54]. The axial components $\lambda_{z,es}$ and $\lambda_{z,gs}$, on the other hand, are found to be zero, consistent with the group theory analysis of C_{3v} systems [90,91]. This leads to the forbidden ISC transition to the spin $|0\rangle$ states.

The nonradiative ISC rates γ between the initial (i) and final (j) states can be expressed by first-order perturbation theory with the Fermi's Golden rule, i.e., $\gamma_{ij} = 4\pi \hbar \lambda^2 \tilde{X}_{ij}$, where $\lambda = \langle i | H_{\text{SOC}} | j \rangle$ is the SOC coupling parameter between states i and j and $\tilde{X}_{ij}$ is the phonon coupling term in Eq. (12) [84]. We further estimate the ISC rate for the spin $|\pm\rangle$ channel using the effective phonon approach as detailed in Sec. XII of the supplemental material [41].

The ISC lifetime (the inverse of the rate) corresponding to $\lambda_{\perp,gs}$ and $\lambda_{\perp,es}$ is found to be $\text{ISC}_{\perp,gs} = 0.25$ ns and $\text{ISC}_{\perp,es} = 95.20$ ps, respectively—both are shorter than the spin-conserved radiative lifetime between triplet states.

In contrast to NV center, the dynamic and pseudo Jahn-Teller (JT) effects are not observed in our system, i.e., the high-symmetry structure remains stable against symmetry-lowering distortion. We attribute this to the strong SOC in the system, which quenches the Jahn-Teller effect and suppresses structural distortion [92,93]. This quenching can be qualitatively understood using a spin-coupled “Mexican triplet hat” Jahn-Teller model, which is derived and discussed in detail in the supplemental material [41], Sec. XIII.

III. DISCUSSION

Based on the spin multiplet structure and excited-state dynamics of the $(\text{MoZnVO})^{2+}$ defect in Fig. 7(a), we next discuss its optical read-out capability as a spin qubit. While $(\text{MoZnVO})^{2+}$ exhibits a similar four-level structure as the NV center in diamond, it offers unique advantages for high-fidelity single-shot spin readout and direct coherent control.

These advantages arise from its significantly stronger SOC compared to the NV center [94–97].

The schematic diagram for single shot readout is shown in Fig. 7(b), where E_x and A_1 refer to transitions between the ground state (GS, 3A_2) and excited state (ES, 3E) within the spin channel $|0\rangle$ and $|+\rangle$, respectively. They consist of optical excitation (red), radiative transition (orange), and nonradiative recombination (blue). The spin-conserved nonradiative processes could be electron-phonon mediated processes as discussed in section D or SOC-mediated ISC paths ($^3E^{|\pm\rangle} \rightarrow ^1A_1 \rightarrow ^1E \rightarrow ^3A_2^{|\pm\rangle}$ for A_1 and $^3E^{(0)} \rightarrow ^1A_1 \rightarrow ^1E \rightarrow ^3A_2^{(0)}$ for E_x). The cross lines represent the spin-flip process, where the spin-flip from $|0\rangle$ of 3E to $|\pm\rangle$ of 3A_2 ($\gamma_{\perp,01}$) could arise from the ISC recombination path of $^3E^{(0)} \rightarrow ^1A_1 \rightarrow ^1E \rightarrow ^3A_2^{|\pm\rangle}$; similarly, the spin-flip from $|\pm\rangle$ ES to $|0\rangle$ GS ($\gamma_{\perp,10}$) could arise from the ISC recombination path of $^3E^{|\pm\rangle} \rightarrow ^1A_1 \rightarrow ^1E \rightarrow ^3A_2^{(0)}$.

The preserved C_{3v} symmetry in the absence of Jahn-Teller distortion leads to spin-selective ISC and results in the following consequences: (a) fast nonradiative recombination in A_1 (solid blue line) due to large $\lambda_{\perp,es/gs}$; (b) forbidden nonradiative recombination in E_x (blue-dashed line) due to $\lambda_{z,gs/es} = 0$; (c) forbidden spin-flip transitions: $\gamma_{\perp,01} = 0$ (caused by $\lambda_{z,es} = 0$) and $\gamma_{\perp,10} = 0$ (caused by $\lambda_{z,gs} = 0$).

First, due to C_{3v} symmetry, the first-order SOC vanishes in 3A_2 but remains finite in the 3E . Combined with strong SOC, this results in a pronounced difference between the ground and excited triplet state spin splitting, which also leads to a large optical transition energy difference, i.e., the energy difference between the E_x and A_1 transitions in Fig. 7(b), enabling resonant excitation control even at high temperatures. Furthermore, the spin-resolved photoluminescence (PL) peaks from these two transitions are inherently distinguishable by intensity: the E_x related optical peak is expected to be significantly brighter than those of A_1 . For the E_x , the fully forbidden nonradiative recombination leading to dominant spin-conserved radiative recombination—hence, strong PL intensity. In contrast, the A_1 transitions include fast nonradiative ISC that competes with radiative recombination, resulting in dimmer PL peaks.

Finally, the forbidden spin-flip transitions of $\gamma_{\perp,01}$ and $\gamma_{\perp,10}$ lead to highly cyclic optical transitions that support high-fidelity single-shot spin readout. Moreover, the readout through the E_x transition is especially robust at elevated temperatures and magnetic fields, because the large spin splitting of the excited state suppresses spin mixing between its sublevels, resulting in minimal $\gamma_{\perp,01}$. Specifically, resonant excitation of the E_x allows for an effectively unlimited number of photon cycles, as $\gamma_{\perp,01}$ remains strictly forbidden for this state. The spin readout fidelity via resonant excitation of E_x can be defined as $F_{00} = 1 - F_{\pm 0}$, where $F_{\pm 0}$ is the probability of obtaining the

measurement outcome $|\pm\rangle$ state after optically pumping into $|0\rangle$ state [31]. The spin-flip probability $p = \frac{\gamma_{\perp,01}}{\gamma_{\perp,01} + \tau_R^{-1}}$, which constitutes the main source of error $F_{\pm 0}$ [29], is also diminished, resulting in high spin readout fidelity. Such high fidelity single-shot readout enables the spin initialization of a single defect center. Once the spin state is read out, arbitrary spin initialization can be performed via radio-frequency (RF) control.

IV. SUMMARY

In conclusion, we have computationally identified $(\text{MoZnVO})^{2+}$ as a deep-level defect qubit in ZnO with the essential ingredients for high-fidelity single-shot readout and long spin coherence time that are robust against environmental fluctuations. Our systematic screening based on thermodynamic stability and optical transition property calculations reveals that $(\text{MoZnVO})^{2+}$ is an exceptional defect, which combines a spin-triplet ground state ($S = 1$), bright visible-range emission with high quantum yield, and an unusually small Huang-Rhys factor (~ 4.9), leading to a sharp zero-phonon line in ZnO. Spin decoherence analysis further reveals long T_2 times approaching 4 ms with a critical threshold paramagnetic impurity concentration of 0.035 ppm that marks the transition from nuclear- to electron-spin-dominated decoherence.

We also examined the electronic spin multiplet structure and dynamics of $(\text{MoZnVO})^{2+}$, including spin-orbit coupling, zero-field splitting, the Jahn-Teller effect, and intersystem crossing. Distinctively, this defect combines strong spin-orbit coupling with the absence of Jahn-Teller distortion, which enables high-fidelity single-shot readout and direct coherent spin control with minimal spin leakage at elevated temperatures. Together, these results establish $(\text{MoZnVO})^{2+}$ as the first deep-level spin qubit candidate in ZnO, opening a pathway toward oxide-based quantum technologies that combine room-temperature operation, photonic integrability, and scalable defect engineering.

V. COMPUTATIONAL METHODS

A. First-principles calculations

We performed the ground-state density-functional theory calculations using the HSE hybrid functional [98], by the plane-wave code Vienna Ab initio Simulation Package (VASP) [99–101]. The multideterminant constrained DFT (mcDFT) was used for geometry relaxations of the excited states. The defect was constructed and relaxed in a $4 \times 4 \times 3$ supercell of ZnO. We use a plane-wave cut-off of 400 eV and the projector-augmented wave (PAW) pseudopotentials to conduct the structural relaxations. We choose the Fock exchange fraction parameter of 0.375 and screening parameter $\omega = 0.2 \text{ \AA}$, which reproduces the experimental lattice constants and band gap of ZnO [102–105]. We obtain a band gap of 3.43 eV, and a lattice

parameter ($a = 3.249 \text{ \AA}$, $c = 5.204 \text{ \AA}$) of the pristine zinc oxide, in good agreement with experimental data (band gap 3.4 eV, lattice constants $a = 3.25 \text{ \AA}$ and $c = 5.21 \text{ \AA}$).

We use the r2SCAN functional for phonon calculations, which is known for its reliability in capturing the static and dynamical properties of the lattice with a low computational cost [106]. We use the Hubbard U correction of 2 eV for the 3d orbital of Zn and obtain the phonon band with very good agreement with experiments (see Fig. S10 [41]).

B. Defect formation energy and charge transition level

The charged defect formation energy $E_f^q(d)$ of the defect d with charge state q is calculated by

$$E_f^q(d) = E_{\text{tot}}^q(d) - E_{\text{tot}}(p) - \sum_i \Delta N_i \mu_i + qE_F + E_{\text{corr}}, \quad (2)$$

where $E_{\text{tot}}^q(d)$ is the total energy of the supercell containing the defect with charge state q and $E_{\text{tot}}(p)$ is the total energy of the pristine system in the same supercell as the defect. ΔN_i denotes the difference in the number of atoms of type i between the defect and pristine systems ($\Delta N_i > 0$ means an atom of type i has been added to the defect system and $\Delta N_i < 0$ means that an atom has been removed). μ_i and E_F are the chemical potential of species i and the Fermi energy, respectively. E_{corr} is charged cell correction to eliminate the fictitious Coulomb interaction of defects with its own periodic images and fictitious homogeneous compensating background [107,108].

In X_{ZnVO} defect complex, the third term in Eq. (2) can be written as $\sum_i \Delta N_i \mu_i = -\mu_{\text{Zn}} - \mu_{\text{O}} + \mu_{\text{X}}$. The formation energy of each defect is calculated with chemical potentials in two conditions: O-rich and O-poor. In the O-rich condition the oxygen chemical potential is computed by total energy of the oxygen molecule ($\mu_{\text{O}}^{\text{O-rich}} = 1/2 E_{\text{tot}}(\text{O}_2)$). In the O-poor condition the zinc chemical potential is computed by $\mu_{\text{Zn}}^{\text{O-poor}} = E_{\text{tot}}(\text{Zn})$, where $E_{\text{tot}}(\text{Zn})$ is the energy of zinc crystal [40]. The $\mu_{\text{O}}^{\text{O-poor}}$ and $\mu_{\text{Zn}}^{\text{O-rich}}$ are computed from $\mu_{\text{Zn}}^{\text{O-poor}}$ and $\mu_{\text{O}}^{\text{O-rich}}$ according to the constraint $\mu_{\text{Zn}}^{\text{O-poor/rich}} + \mu_{\text{O}}^{\text{O-poor/rich}} = \mu_{\text{ZnO}}$. The chemical potential of the dopant X ($\text{X} = \text{Ti}, \text{Nb}, \text{V}, \text{Mo}$) is computed from its most stable oxide compound, where μ_{Ti} , μ_{Nb} , μ_{V} , and μ_{Mo} are computed from TiO_2 , Nb_2O_5 , V_2O_3 , and MoO_3 , respectively [109].

The thermodynamic charge transition level (CTL) between charge states q and q' ($\epsilon(q/q')$) is the Fermi-level position at which the formation energy of charge state q and q' are the same [108,109]:

$$\epsilon(q/q') = \frac{E_f^q(E_F = 0) - E_f^{q'}(E_F = 0)}{q' - q}. \quad (3)$$

Here, $E_f^q(E_F = 0)$ is the defect formation energy of the defect at the Fermi level $E_F = 0$ (aligned with VBM). The defect system is more stable at a charge state q when its Fermi level is smaller than CTL $\epsilon(q/q')$ and is more stable at q' when $E_F > \epsilon(q/q')$.

C. Absorption spectrum and radiative lifetime

The optical absorption spectrum at the random-phase approximation (RPA) is computed with the Yambo code [46], including the local field effect for the polarizability, with the input single particle states from hybrid functional HSE (Fock exchange = 0.375) [110]. The DFT single particle states are calculated by the open source plan-wave code Quantum Espresso [111] with norm-conserving Vanderbilt (ONCV) pseudopotentials [112,113] and wavefunction cutoff of 80 Ry. We then extract the transition dipole moment and oscillator strength for each excitation. The radiative lifetime for defects transition is derived from the Fermi's golden rule [84,114] and computed by

$$\tau_R = \frac{3\pi\epsilon_0\hbar^4 c^3}{n_D E^2 \mu^2}, \quad (4)$$

where E is the excitation energy, c is the speed of light, μ^2 is the modulus square of transition dipole moment, and $n_D = \sqrt{\epsilon} = 2.4$ is the reflective index computed from the dielectric constant of pristine ZnO.

D. Multideterminant cDFT

The idea of multideterminant cDFT is that the energy expectation value of a linear combination of states is a linear combination of the energies of the pure states. This idea has been employed on describing the multireference nature for spin defects in past works [115–119]. For the $(\text{Mo}_{\text{ZnVO}})^{2+}$ and $(\text{Nb}_{\text{ZnVO}})^+$, the two-particle wavefunction constructed from the two highest occupied $|e_{x/y}\rangle$ orbitals can be written as

$$|{}^3A_2^{m_s=1}\rangle = \frac{1}{\sqrt{2}}(|e_x e_y\rangle - |e_y e_x\rangle), \quad (5a)$$

$$|{}^3A_2^{m_s=0}\rangle = \frac{1}{\sqrt{2}}(|e_x \bar{e}_y\rangle - |e_y \bar{e}_x\rangle), \quad (5b)$$

$$|{}^3A_2^{m_s=-1}\rangle = \frac{1}{\sqrt{2}}(|\bar{e}_x \bar{e}_y\rangle - |\bar{e}_y \bar{e}_x\rangle), \quad (5c)$$

$$|{}^1E_x\rangle = \frac{1}{\sqrt{2}}(|e_x \bar{e}_y\rangle + |e_y \bar{e}_x\rangle), \quad (5d)$$

$$|{}^1E_y\rangle = \frac{1}{\sqrt{2}}(|e_x \bar{e}_x\rangle - |e_y \bar{e}_y\rangle), \quad (5e)$$

where “ $\bar{e}_{x/y}$ ” represent the occupied spin minority $e_{x/y}$ state and “ $e_{x/y}$ ” represent the occupied spin majority $e_{x/y}$. The triplet ground states $|{}^3A_2^{m_s=\pm 1}\rangle$ and $|{}^3A_2^{m_s=0}\rangle$

should have degenerate energy $E(|^3A_2\rangle)$, we therefore use the single configuration at $m_s = 1$ to compute the ground-state energy: $E(|^3A_2\rangle) = E(|e_x e_y\rangle)$. The degenerated triplet excited state $^3E_{x/y}$ (correspond to the spin majority $e \rightarrow a_1$ transition) can also be computed from the single determinant: $E(|^3E_{x/y}\rangle) = E(|2a_1 e_{x/y}\rangle)$. In the calculation, we use the fractional occupation $E(|2a_1 e\rangle) = E(|2a_1 e_x^{0.5} e_y^{0.5}\rangle)$ to preserve the state degeneracy. The $E(|^3A'_2\rangle) = E(|\bar{a}_1 e_x e_y 2\bar{a}_1\rangle)$ state corresponds to an $a_1 \rightarrow 2a_1$ excitation and is essentially single-determinantal in character, allowing it to be treated directly within a standard cDFT framework.

The DFT close shell calculation for the $(\text{MoZnVO})^{2+}$ broke the degeneracy of the $e_{x/y}$ orbitals, resulting in the spin-symmetry-broken determinant [119]: $|e_- \bar{e}_+\rangle$ and the corresponded energy $E(e_- \bar{e}_+)$ where $e_+ = (e_x + e_y)/\sqrt{2}$ and $e_- = (e_x - e_y)/\sqrt{2}$.

Notice $|e_- \bar{e}_+\rangle = \frac{1}{2} |(e_x - e_y)(\bar{e}_x + \bar{e}_y)\rangle = \frac{1}{\sqrt{2}} (|^1E_y\rangle + |^3A_2^{m_s=0}\rangle)$. We therefore have

$$E(e_- \bar{e}_+) = \langle e_- \bar{e}_+ | H | e_- \bar{e}_+ \rangle \quad (6a)$$

$$= \frac{1}{2} (E(^1E_y) + E(^3A_2^{m_s=0}))$$

$$\Rightarrow E(|^1E\rangle) = 2E(|e_- \bar{e}_+\rangle) - E(|e_x e_y\rangle). \quad (6b)$$

E. Nonradiative lifetime, photoluminescence, and Huang Rhy's factor

The nonradiative lifetime (τ^{NR}) is a measure of how fast the nonradiative recombination happens between the final state $|f\rangle$ and initial state $|i\rangle$. The phonon-assisted nonradiative recombination is evaluated via Fermi's golden rule as below:

$$\frac{1}{\tau_{if}^{NR}} = \frac{2\pi}{\hbar} g \sum_{n,m} p_{in} |\langle fm | H^{e-ph} | in \rangle|^2 \delta(E_{fm} - E_{in}), \quad (7)$$

where H^{e-ph} is the electron-phonon coupling Hamiltonian, g is the degeneracy factor of the final state that depends on the number of equivalent atomic configurations, and p_{in} is the occupation number of the vibronic state $|in\rangle$ following the Boltzmann distribution.

Under the static coupling approximation with one-dimensional (1D) phonon approximation [58,84], we can

rewrite Eq. (7) as

$$\frac{1}{\tau_{if}^{NR}} = \frac{2\pi}{\hbar} g |W_{if}|^2 X_{if}(T) \quad (8)$$

$$W_{if} = \langle \psi_i(\mathbf{r}, \mathbf{R}) | \frac{\partial H}{\partial \mathbf{Q}} | \psi_f(\mathbf{r}, \mathbf{R}) \rangle_{\mathbf{R}=\mathbf{R}_a} \quad (9)$$

$$X_{if} = \sum_{n,m} p_{in} |\langle \phi_{fm}(\mathbf{R}) | Q - Q_a | \phi_{in}(\mathbf{R}) \rangle|^2 \times \delta(m\hbar\omega_f - n\hbar\omega_i + \Delta E_{if}). \quad (10)$$

Equation (8) is separated into the electronic term (W_{if}), which depends on the electronic wave function (ψ) overlap, and the phonon term (X_{if}), which describes the strength of the phonon contribution. W_{if} is determined using finite differences of Kohn-Sham orbitals from DFT calculation using HSE (0.375) functional. The phonon term X_{if} includes the energy conservation between the initial and final vibronic states with vibrational frequencies of ω_i and ω_f , and ϕ is the phonon wave function obtained from harmonic oscillator wavefunctions. The detailed derivation can be found in Refs. [58,60].

To validate the 1D phonon approximation, we compare the Huang-Rhys factor calculated with the 1D effective phonon and full phonon results, as detailed in Table S8 [41]. To compute the intersystem crossing (ISC) rate, we adopted the approach derived from nonradiative rates, as implemented in our in-house code [54,84],

$$\Gamma_{ISC} = 4\pi\hbar\lambda_{\perp}^2 \tilde{X}_{if}(T), \quad (11)$$

$$\tilde{X}_{if}(T) = \sum_{n,m} p_{in} |\langle \phi_{fm}(\mathbf{R}) | \phi_{in}(\mathbf{R}) \rangle|^2 \times \delta(m\hbar\omega_f - n\hbar\omega_i + \Delta E_{if}). \quad (12)$$

This method allows different values for the initial-state vibrational frequency (ω_i) and the final-state frequency (ω_f) through explicit calculations of the phonon wavefunction overlap, and we obtain good agreement with experimental values of ISC rates for NV center in diamond [54,84].

The photoluminescence lineshape spectrum was simulated using the Huang-Rhys method with all phonon eigenmodes, implemented in our in-house codes [61,69], with $\gamma = 0.005$ eV and smearing = 0.003 eV, where γ is a free parameter that accounts for the broadening of PL. Due to the well-known failure of local and semilocal exchange correlation functional on the ZnO system [37,120] and the high computational cost of the hybrid functional, we apply the r2SCAN functional with Hubbard U for the all phonon calculations using the Phonopy code interfaced with VASP and Hubbard U corrections of $U_{Zn-d} = 2$ eV and $U_{Nb-d} = 3.7$ eV. It has been tested that this method reproduces the defect local structure comparable to HSE as well as the phonon band of pristine ZnO (see Fig. S10 [41]).

F. Decoherence time

G. Quantum bath model to compute the spin decoherence

We employ the quantum bath theory to compute the spin decoherence [121–123], in which the decoherence occurs due to the entanglement between a central spin and its environment. We consider electronic and nuclear spin baths as the environment. Bath spins are randomly distributed in the lattice and bath spins, within a certain radius from the defect qubit, are included in the calculation. This bath radius (r_{bath}) is determined by performing a systematic convergence test as shown in Fig. S12 [41]. We find that a radius of 5 nm gives a numerically converged result for the nuclear spin bath. For the electronic spin bath, we find that the bath radii of 16, 37, 85, and 220 nm are appropriate for 100, 10, 1, and 0.1 ppm of electronic spin concentrations, respectively.

In addition, we use another parameter (r_{dipole}), which sets the maximum distance for the interaction between bath spins. This means that if two bath spins are separated by a distance larger than r_{dipole} , the two spins are considered noninteracting. We find that our CCE calculations are converged with r_{dipole} of 1 nm for the nuclear spin bath, and for the electronic spin bath, r_{dipole} of 13, 33, 62, and 120 nm for 100, 10, 1, and 0.1 ppm of electronic spin concentrations.

The dynamics of the total system of qubit and environment is governed by a spin Hamiltonian, which is expressed as

$$H = H_d + H_{\text{bath}} + H_{d-\text{bath}}, \quad (13)$$

where H_d is the qubit Hamiltonian, H_{bath} is the bath spin Hamiltonian, and $H_{d-\text{bath}}$ is the interaction between the qubit and the bath spins. For the nuclear spin bath, each component of the Hamiltonian is given as follows:

$$\begin{aligned} H_d &= -\gamma_e \mathbf{B} \cdot \mathbf{S}, \\ H_{\text{bath}} &= -\mathbf{B} \cdot \sum_i \gamma_{n_i} \mathbf{I}_i + H_{n-n} + H_Q, \\ H_{d-\text{bath}} &= S_z \sum_i \mathbf{A}_i \cdot \mathbf{I}_i \\ &= \sum_i (B_{ix} I_{ix} S_z + B_{iy} I_{iy} S_z + A_i I_{iz} S_z), \\ H_{n-n} &= \frac{\mu_0}{4\pi} \sum_{i,j} \gamma_{n_i} \gamma_{n_j} \left[\frac{\mathbf{I}_i \cdot \mathbf{I}_j}{r_{ij}^3} - \frac{3(\mathbf{I}_i \cdot \mathbf{r}_{ij})(\mathbf{I}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5} \right], \\ H_Q &= \frac{eQ}{6I(2I-1)} \sum_{\alpha,\beta} V_{\alpha\beta} \left[\frac{3}{2} (I_\alpha I_\beta + I_\beta I_\alpha) - \delta_{\alpha\beta} \mathbf{I}^2 \right]. \end{aligned} \quad (14)$$

In these equations, γ_e and γ_{n_i} are the gyromagnetic ratios of the electron spin ($\mathbf{S}$) and the i th nuclear spin ($\mathbf{I}_i$) in the

bath, respectively. The external magnetic field is aligned parallel to the defect's symmetry axis. We adopt the secular approximation for the hyperfine interaction ($H_{d-\text{bath}}$), in which the nonsecular terms including S_x and S_y are neglected. In the nuclear spin-spin interaction (H_{n-n}), μ_0 is the vacuum permeability and r_{ij} is the distance between the i th nuclear spin and the j th nuclear spin.

H_Q is the nuclear quadrupole interaction, in which eQ is the quadrupole moment of the isotope under consideration interacting with the EFG tensor $V_{\alpha\beta}$ ($\alpha, \beta = x, y, z$).

The hyperfine tensor ($\mathbf{A}$) and the electric field gradient (EFG) tensor ($V_{\alpha\beta}$) are computed by using DFT as implemented in Vienna Ab initio Simulation Package (VASP) code at HSE hybrid functional level of theory.

The spin Hamiltonian for the electronic spin bath can be expressed as follows:

$$\begin{aligned} H_d &= -\gamma_e \mathbf{B} \cdot \mathbf{S}, \\ H_{\text{bath}} &= -\mathbf{B} \sum_i \gamma_{e_i} \mathbf{S}_i + H_{e-e}, \\ H_{e-e} &= -\frac{\mu_0}{4\pi} \sum_{i,j} \gamma_{e_i} \gamma_{e_j} \left[\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{r_{ij}^3} - \frac{3(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5} \right], \end{aligned} \quad (15)$$

Similar to the case of the nuclear spin bath, the qubit Hamiltonian H_d represents the Zeeman interaction of the qubit (electron spin) with an external magnetic field $\mathbf{B}$. Here, γ_e is the gyromagnetic ratio of the electron and S_z is the z component of the electron spin operator. The bath spin Hamiltonian describes the dynamics of the bath spins (other electron spins in the environment). The first term of the bath spin Hamiltonian represents the Zeeman interaction of each electron spin in the bath with the external magnetic field. H_{e-e} in the bath spin Hamiltonian represents the magnetic dipolar interactions between the electron spins in the bath. Here, $\mathbf{S}_i$ represents the spin operator of the i th electronic spin in the bath.

The coherence function $\mathcal{L}(t)$ is given as the off-diagonal element of the reduced density matrix, formally expressed as

$$\mathcal{L}(t) \equiv \frac{\text{tr}[\rho_{\text{tot}}(t) S_+]}{\text{tr}[\rho_{\text{tot}}(0) S_+]}, \quad (16)$$

where $\rho_{\text{tot}}(t)$ is the total density matrix of the qubit (ρ_e) and the bath (ρ_{bath}) at time t . S_+ is the electron spin raising operator, defined as $S_+ = S_x + iS_y$. To compute the coherence function, we employ the cluster correlation expansion (CCE) technique [124], which enables a systematic expansion of the coherence function in many-body systems. We find that second-order CCE (CCE-2) and first-order CCE (CCE-1) give numerically converged results for the Hahn-echo decay time (T_2) and the FID time (T_2^*), respectively.

H. ZFS and SOC strength

The ZFS parameters (D and E) consist of first-order spin-spin interaction contributions (D_{SS}, E_{SS}) and second-order spin-orbit (SO) contributions (D_{SOC}, E_{SOC}) [86]. For transition metals, the SO contribution is significant and cannot be neglected. We computed the SO contribution to ZFS using the linear response method implemented in ORCA [125,126], employing all-electron PBE0 calculations with the def2-TZVP basis set [127]. In contrast, the spin-spin contribution to ZFS, which is relatively long-ranged compared to the SO contribution, did not converge with the cluster size (see Table S10 [41]). To address this, we calculated the spin-spin contribution using our in-house code [84] interfaced with the plane-wave code QUANTUM ESPRESSO [111].

We use the complete-active-space self-consistent field (CASSCF) method implemented in the ORCA code [125,126] to obtain SOC matrix elements between the electronic states. The second-order Douglas-Kroll-Hess (DKH2) Hamiltonian [128], the all-electron DKH-def2-TZVP basis set [129], and the SARC-DKH-TZVP basis set [130] are used to account for the scalar relativistic effects. The spin-orbit mean-field operator is used for the SOC calculation [131]. The cluster is created by retaining atoms near the defects and passivate the surface dangling bond with pseudo hydrogen atoms with core charge of $q=(8-m)/4$ [132], where we choose the O-H bond to be 1.057 Å and Zn-H to be 1.731 Å [133]. The perturbative correction with N-electron Valence State Perturbation Theory (NEVPT2) [51] was applied upon the CASSCF to obtain the multireference excited state with both static and dynamic correlation. More details can be found in supplemental material, Sec. XI [41].

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The authors declare no competing interests.

CODE AVAILABILITY

The codes for electronic structure are available through open-source software QUANTUM ESPRESSO [111], the WEST-code [45], and the ORCA code. The in-house open source codes for calculating nonradiative and intersystem crossing rates [11,58,84] are available from GitHub <https://github.com/Ping-Group-UCSC/NonRad-ISC>.

DATA AVAILABILITY

The data that support the findings of this article are openly available [134], embargo periods may apply.

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- [1] M. W. Doherty, N. B. Manson, P. Delaney, F. Jelezko, J. Wrachtrup, and L. C. L. Hollenberg, The nitrogen-vacancy colour centre in diamond, *Phys. Rep.* **528**, 1 (2013).
 - [2] L. Orphal-Kobin, C. G. Torun, J. M. Bopp, G. Pieplow, and T. Schröder, Coherent microwave, optical, and mechanical quantum control of spin qubits in diamond, *Adv. Quantum Technol.* **8**, 2300432 (2025).
 - [3] I. B. W. Harris, and D. Englund, Coherence of group-IV color centers, *Phys. Rev. B* **109**, 085414 (2024).

- [4] I. Karapatzakis, J. Resch, M. Schrodin, P. Fuchs, M. Kieschnick, J. Heupel, L. Kussi, C. Sürgers, C. Popov, J. Meijer, C. Becher, W. Wernsdorfer, and D. Hunger, Microwave control of the tin-vacancy spin qubit in diamond with a superconducting waveguide, *Phys. Rev. X* **14**, 031036 (2024).
- [5] S. A. Tarasenko, A. V. Poshakinskiy, D. Simin, V. A. Soltamov, E. N. Mokhov, P. G. Baranov, V. Dyakonov, and G. V. Astakhov, Spin and optical properties of silicon vacancies in silicon carbide—A review, *Phys. Status Solidi B* **255**, 1700258 (2018).
- [6] S. Castelletto, and A. Boretti, Silicon carbide color centers for quantum applications, *J. Phys. Photonics* **2**, 022001 (2020).
- [7] A. Gottscholl, M. Kianinia, V. Soltamov, S. Orlinskii, G. Mamin, C. Bradac, C. Kasper, K. Krambrock, A. Sperlich, M. Toth, I. Aharonovich, and V. Dyakonov, Initialization and read-out of intrinsic spin defects in a van der Waals crystal at room temperature, *Nat. Mater.* **19**, 540 (2020).
- [8] A. Gottscholl, M. Diez, V. Soltamov, C. Kasper, A. Sperlich, M. Kianinia, C. Bradac, I. Aharonovich, and V. Dyakonov, Room temperature coherent control of spin defects in hexagonal boron nitride, *Sci. Adv.* **7**, eabf3630 (2021).
- [9] R. Gong, G. He, X. Gao, P. Ju, Z. Liu, B. Ye, E. A. Henriksen, T. Li, and C. Zu, Coherent dynamics of strongly interacting electronic spin defects in hexagonal boron nitride, *Nat. Commun.* **14**, 3299 (2023).
- [10] S. Kanai, F. J. Heremans, H. Seo, G. Wolfowicz, C. P. Anderson, S. E. Sullivan, M. Onizhuk, G. Galli, D. D. Awschalom, and H. Ohno, Generalized scaling of spin qubit coherence in over 12,000 host materials, *Proc. Natl. Acad. Sci. U.S.A.* **119**, e2121808119 (2022).
- [11] Y. Ping, and T. J. Smart, Computational design of quantum defects in two-dimensional materials, *Nat. Comput. Sci.* **1**, 646 (2021).
- [12] D. D. Awschalom, R. Hanson, J. Wrachtrup, and B. B. Zhou, Quantum technologies with optically interfaced solid-state spins, *Nat. Photonics* **12**, 516 (2018).
- [13] J. R. Weber, W. F. Koehl, J. B. Varley, A. Janotti, B. B. Buckley, C. G. Van De Walle, and D. D. Awschalom, Quantum computing with defects, *Proc. Natl. Acad. Sci. U.S.A.* **107**, 8513 (2010).
- [14] J. Falson, D. Maryenko, Y. Kozuka, A. Tsukazaki, and M. Kawasaki, Magnesium doping controlled density and mobility of two-dimensional electron gas in $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$ heterostructures, *Appl. Phys. Express* **4**, 091101 (2011).
- [15] Q. Li, J. Zhang, J. Chong, and X. Hou, Electron mobility in ZnMgO/ZnO heterostructures in the Bloch–Grüneisen regime, *Appl. Phys. Express* **6**, 121102 (2013).
- [16] J. Falson, Y. Kozuka, M. Uchida, J. H. Smet, T.-h. Arima, A. Tsukazaki, and M. Kawasaki, MgZnO/ZnO heterostructures with electron mobility exceeding $1 \times 10^6 \text{ cm}^2/\text{Vs}$, *Sci. Rep.* **6**, 26598 (2016).
- [17] W. Haynes, *CRC Handbook of Chemistry and Physics* (Taylor & Francis Group, Boca Raton, FL, 2010), 91st ed.
- [18] V. Niaouris, M. V. Durnev, X. Linpeng, M. L. K. Viitaniemi, C. Zimmermann, A. Vishnuradhan, Y. Kozuka, M. Kawasaki, and K.-M. C. Fu, Ensemble spin relaxation of shallow donor qubits in ZnO, *Phys. Rev. B* **105**, 195202 (2022).
- [19] X. Linpeng, M. L. Viitaniemi, A. Vishnuradhan, Y. Kozuka, C. Johnson, M. Kawasaki, and K.-M. C. Fu, Coherence properties of shallow donor qubits in ZnO, *Phys. Rev. Appl.* **10**, 064061 (2018).
- [20] X. Wang, C. Zimmermann, M. Titze, V. Niaouris, E. R. Hansen, S. H. D'Ambrosia, L. Vines, E. S. Bielejec, and K.-M. C. Fu, Properties of donor qubits in ZnO formed by indium-ion implantation, *Phys. Rev. Appl.* **19**, 054090 (2023).
- [21] M. L. K. Viitaniemi, C. Zimmermann, V. Niaouris, S. H. D'Ambrosia, X. Wang, E. S. Kumar, F. Mohammadbeigi, S. P. Watkins, and K.-M. C. Fu, Coherent spin preparation of indium donor qubits in single ZnO nanowires, *Nano Lett.* **22**, 2134 (2022).
- [22] V. Niaouris, S. H. D'Ambrosia, C. Zimmermann, X. Wang, E. R. Hansen, M. Titze, E. S. Bielejec, and K.-M. C. Fu, Contributions to the optical linewidth of shallow donor-bound excitonic transition in ZnO, *Opt. Quantum* **2**, 7 (2024).
- [23] C. Gray, L. Trefflich, R. Röder, C. Ronning, M. O. Henry, and E. McGlynn, Local atomic environment of the correlated defect in zinc oxide, *J. Phys. D: Appl. Phys.* **50**, 145105 (2017).
- [24] Y.-M. Hao, S.-Y. Lou, S.-M. Zhou, R.-J. Yuan, G.-Y. Zhu, and N. Li, Structural, optical, and magnetic studies of manganese-doped zinc oxide hierarchical microspheres by self-assembly of nanoparticles, *Nanoscale Res. Lett.* **7**, 100 (2012).
- [25] S.-C. Liao, H.-F. Lin, S.-W. Hung, and C.-T. Hu, DC thermal plasma synthesis and properties of zinc oxide nanorods, *JVST B* **24**, 1322 (2006).
- [26] H. Shahroosvand, and M. Ghorbani-asl, Solution-based synthetic strategies for EU doped ZnO nanoparticle with enhanced red photoluminescence, *J. Lumin.* **144**, 223 (2013).
- [27] S. Singh, D. Nakamura, K. Sakai, T. Okada, and M. S. Ramachandra Rao, Investigation of low-temperature excitonic and defect emission from Ni-doped ZnO nanoneedles and V-doped ZnO nanostructured film, *New J. Phys.* **12**, 023007 (2010).
- [28] D. M. Irber, F. Poggiali, F. Kong, M. Kieschnick, T. Lühmann, D. Kwiatkowski, J. Meijer, J. Du, F. Shi, and F. Reinhard, Robust all-optical single-shot readout of nitrogen-vacancy centers in diamond, *Nat. Commun.* **12**, 532 (2021).
- [29] D. J. Christle, P. V. Klimov, C. F. De Las Casas, K. Szász, V. Ivády, V. Jokubavicius, J. Ul Hassan, M. Syväjärvi, W. F. Koehl, T. Ohshima, N. T. Son, E. Janzén, Á. Gali, and D. D. Awschalom, Isolated spin qubits in SiC with a high-fidelity infrared spin-to-photon interface, *Phys. Rev. X* **7**, 021046 (2017).
- [30] D. Hopper, H. Shulevitz, and L. Bassett, Spin readout techniques of the nitrogen-vacancy center in diamond, *Micromachines* **9**, 437 (2018).
- [31] L. Robledo, L. Childress, H. Bernien, B. Hensen, P. F. A. Alkemade, and R. Hanson, High-fidelity projective readout of a solid-state spin quantum register, *Nature* **477**, 574 (2011).

- [32] E. I. Rosenthal, S. Biswas, G. Scuri, H. Lee, A. J. Stein, H. C. Kleidermacher, J. Grzesik, A. E. Rugar, S. Aghaeimeibodi, D. Riedel, M. Titze, E. S. Bielejec, J. Choi, C. P. Anderson, and J. Vučković, Single-shot readout and weak measurement of a tin-vacancy qubit in diamond, *Phys. Rev. X* **14**, 041008 (2024).
- [33] C. P. Anderson, E. O. Glen, C. Zeledon, A. Bourassa, Y. Jin, Y. Zhu, C. Vorwerk, A. L. Crook, H. Abe, J. Ul-Hassan, T. Ohshima, N. T. Son, G. Galli, and D. D. Awschalom, Five-second coherence of a single spin with single-shot readout in silicon carbide, *Sci. Adv.* **8**, eabm5912 (2022).
- [34] W. F. Koehl, B. B. Buckley, F. J. Heremans, G. Calusine, and D. D. Awschalom, Room temperature coherent control of defect spin qubits in silicon carbide, *Nature* **479**, 84 (2011).
- [35] D. D. Sukachev, A. Sipahigil, C. T. Nguyen, M. K. Bhaskar, R. E. Evans, F. Jelezko, and M. D. Lukin, Silicon-vacancy spin qubit in diamond: A quantum memory exceeding 10 ms with single-shot state readout, *Phys. Rev. Lett.* **119**, 223602 (2017).
- [36] H. Seo, H. Ma, M. Govoni, and G. Galli, Designing defect-based qubit candidates in wide-gap binary semiconductors for solid-state quantum technologies, *Phys. Rev. Mater.* **1**, 075002 (2017).
- [37] F. Oba, M. Choi, A. Togo, and I. Tanaka, Point defects in ZnO: An approach from first principles, *STAM* **12**, 034302 (2011).
- [38] Y. K. Frodason, K. M. Johansen, T. S. Bjørheim, B. G. Svensson, and A. Alkauskas, Zn vacancy as a polaronic hole trap in ZnO, *Phys. Rev. B* **95**, 094105 (2017).
- [39] S. J. Clark, J. Robertson, S. Lany, and A. Zunger, Intrinsic defects in ZnO calculated by screened exchange and hybrid density functionals, *Phys. Rev. B* **81**, 115311 (2010).
- [40] A. Janotti, and C. G. Van De Walle, Native point defects in ZnO, *Phys. Rev. B* **76**, 165202 (2007).
- [41] See supplemental material at <http://link.aps.org/supplemental/10.1103/v3fp-821b> for additional discussion, analysis, and numerical data.
- [42] M. D. McCluskey, and S. J. Jokela, Defects in ZnO, *J. Appl. Phys.* **106**, 071101 (2009).
- [43] L. Liu, Z. Mei, A. Tang, A. Azarov, A. Kuznetsov, Q.-K. Xue, and X. Du, Oxygen vacancies: The origin of n-type conductivity in ZnO, *Phys. Rev. B* **93**, 235305 (2016).
- [44] A. Tsukazaki, H. Yuji, S. Akasaka, K. Tamura, K. Nakahara, T. Tanabe, H. Takasu, A. Ohtomo, and M. Kawasaki, High electron mobility exceeding $10^4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$ single heterostructures grown by molecular beam epitaxy, *Appl. Phys. Express* **1**, 055004 (2008).
- [45] M. Govoni, and G. Galli, Large scale GW calculations, *J. Chem. Theory Comput.* **11**, 2680 (2015).
- [46] G. Onida, L. Reining, and A. Rubio, Electronic excitations: Density-functional versus many-body Green's-function approaches, *Rev. Mod. Phys.* **74**, 601 (2002).
- [47] H. Ma, N. Sheng, M. Govoni, and G. Galli, First-principles studies of strongly correlated states in defect spin qubits in diamond, *Phys. Chem. Chem. Phys.* **22**, 25522 (2020).
- [48] H. Ma, M. Govoni, and G. Galli, Quantum simulations of materials on near-term quantum computers, *npj Comput. Mater.* **6**, 1 (2020).
- [49] N. Sheng, C. Vorwerk, M. Govoni, and G. Galli, Green's function formulation of quantum defect embedding theory, *J. Chem. Theory Comput.* **18**, 3512 (2022).
- [50] C. Vorwerk, N. Sheng, M. Govoni, B. Huang, and G. Galli, Quantum embedding theories to simulate condensed systems on quantum computers, *Nat. Comput. Sci.* **2**, 424 (2022).
- [51] Y. Guo, K. Sivalingam, E. F. Valeev, and F. Neese, SparseMaps—A systematic infrastructure for reduced-scaling electronic structure methods. III. Linear-scaling multireference domain-based pair natural orbital N-electron valence perturbation theory, *J. Chem. Phys.* **144**, 094111 (2016).
- [52] Y. Ma, M. Rohlfing, and A. Gali, Excited states of the negatively charged nitrogen-vacancy color center in diamond, *Phys. Rev. B* **81**, 041204(R) (2010).
- [53] V. Ivády, G. Barcza, G. Thiering, S. Li, H. Hamdi, J.-P. Chou, Ö. Legeza, and A. Gali, Ab initio theory of the negatively charged boron vacancy qubit in hexagonal boron nitride, *npj Comput. Mater.* **6**, 1 (2020).
- [54] K. Li, V. D. Dergachev, I. D. Dergachev, S. Zhang, S. A. Varganov, and Y. Ping, Excited-state dynamics and optically detected magnetic resonance of solid-state spin defects from first principles, *Phys. Rev. B* **110**, 184302 (2024).
- [55] J. R. Reimers, J. Shen, M. Kianinia, C. Bradac, I. Aharonovich, M. J. Ford, and P. Piecuch, Photoluminescence, photophysics, and photochemistry of the VB^- defect in hexagonal boron nitride, *Phys. Rev. B* **102**, 144105 (2020).
- [56] J. J. Markham, Interaction of normal modes with electron traps, *Rev. Mod. Phys.* **31**, 956 (1959).
- [57] J. Walker, Optical absorption and luminescence in diamond, *Rep. Prog. Phys.* **42**, 1605 (1979).
- [58] F. Wu, T. J. Smart, J. Xu, and Y. Ping, Carrier recombination mechanism at defects in wide band gap two-dimensional materials from first principles, *Phys. Rev. B* **100**, 081407(R) (2019).
- [59] J. L. Lyons, J. B. Varley, D. Steiauf, A. Janotti, and C. G. Van de Walle, First-principles characterization of native-defect-related optical transitions in ZnO, *J. Appl. Phys.* **122**, 035704 (2017).
- [60] A. Alkauskas, Q. Yan, and C. G. Van de Walle, First-principles theory of nonradiative carrier capture via multiphonon emission, *Phys. Rev. B* **90**, 075202 (2014).
- [61] A. Alkauskas, J. L. Lyons, D. Steiauf, and C. G. Van De Walle, First-principles calculations of luminescence spectrum line shapes for defects in semiconductors: The example of GaN and ZnO, *Phys. Rev. Lett.* **109**, 267401 (2012).
- [62] J. L. Lyons, A. Alkauskas, A. Janotti, and C. G. Van De Walle, Deep donor state of the copper acceptor as a source of green luminescence in ZnO, *Appl. Phys. Lett.* **111**, 042101 (2017).
- [63] Y. K. Frodason, K. M. Johansen, T. S. Bjørheim, B. G. Svensson, and A. Alkauskas, Zn vacancy-donor impurity complexes in ZnO, *Phys. Rev. B* **97**, 104109 (2018).

- [64] A. Alkauskas, B. B. Buckley, D. D. Awschalom, and C. G. V. de Walle, First-principles theory of the luminescence lineshape for the triplet transition in diamond NV centres, *New J. Phys.* **16**, 073026 (2014).
- [65] O. Bulancea-Lindvall, J. Davidsson, R. Armiento, and I. A. Abrikosov, Chlorine vacancy in 4 H-SiC: An NV-like defect with telecom-wavelength emission, *Phys. Rev. B* **108**, 224106 (2023).
- [66] M. Mackoito-Sinkevičienė, M. Maciaszek, C. G. Van de Walle, and A. Alkauskas, Carbon dimer defect as a source of the 4.1 eV luminescence in hexagonal boron nitride, *Appl. Phys. Lett.* **115**, 212101 (2019).
- [67] S. A. Tawfik, S. Ali, M. Fronzi, M. Kianinia, T. T. Tran, C. Stampfl, I. Aharonovich, M. Toth, and M. J. Ford, First-principles investigation of quantum emission from hBN defects, *Nanoscale* **9**, 13575 (2017).
- [68] R. D. Shannon, Revised effective ionic radii and systematic studies of interatomic distances in halides and chalcogenides, *Acta Cryst. A* **32**, 751 (1976).
- [69] K. Li, T. J. Smart, and Y. Ping, Carbon trimer as a 2 eV single-photon emitter candidate in hexagonal boron nitride: A first-principles study, *Phys. Rev. Mater.* **6**, L042201 (2022).
- [70] N. Zhao, S.-W. Ho, and R.-B. Liu, Decoherence and dynamical decoupling control of nitrogen vacancy center electron spins in nuclear spin baths, *Phys. Rev. B* **85**, 115303 (2012).
- [71] J. R. Maze, A. Dréau, V. Waselowski, H. Duarte, J.-F. Roch, and V. Jacques, Free induction decay of single spins in diamond, *New J. Phys.* **14**, 103041 (2012).
- [72] R. Nagy, M. Niethammer, M. Widmann, Y.-C. Chen, P. Udvarhelyi, C. Bonato, J. U. Hassan, R. Karhu, I. G. Ivanov, N. T. Son, J. R. Maze, T. Ohshima, Ö. O. Soykal, Á. Gali, S.-Y. Lee, F. Kaiser, and J. Wrachtrup, High-fidelity spin and optical control of single silicon-vacancy centres in silicon carbide, *Nat. Commun.* **10**, 1954 (2019).
- [73] B. C. Rose, C. D. Weis, A. M. Tyryshkin, T. Schenkel, and S. A. Lyon, Spin coherence and 14 N ESEEM effects of nitrogen-vacancy centers in diamond with X-band pulsed ESR, *Diam. Relat. Mater.* **72**, 32 (2017).
- [74] W. B. Mims, Envelope modulation in spin-echo experiments, *Phys. Rev. B* **5**, 2409 (1972).
- [75] L.-P. Yang, C. Burk, M. Widmann, S.-Y. Lee, J. Wrachtrup, and N. Zhao, Electron spin decoherence in silicon carbide nuclear spin bath, *Phys. Rev. B* **90**, 241203(R) (2014).
- [76] O. Bulancea-Lindvall, M. T. Eiles, N. T. Son, I. A. Abrikosov, and V. Ivády, Isotope-purification-induced reduction of spin-relaxation and spin-coherence times in semiconductors, *Phys. Rev. Appl.* **19**, 064046 (2023).
- [77] L. S. Vlasenko, and G. D. Watkins, Optical detection of electron paramagnetic resonance in room-temperature electron-irradiated ZnO, *Phys. Rev. B* **71**, 125210 (2005).
- [78] X. Zuo, S.-D. Yoon, A. Yang, W.-H. Duan, C. Vittoria, and V. G. Harris, Ferromagnetism in pure wurtzite zinc oxide, *J. Appl. Phys.* **105**, 07C508 (2009).
- [79] A. Fedorov, M. Visotin, A. Kholobina, A. Kuzubov, N. Mikhaleva, and H. S. Hsu, Investigation of intrinsic defect magnetic properties in wurtzite ZnO materials, *J. Magn. Magn. Mater.* **440**, 5 (2017).
- [80] Q. L. Lin, G. P. Li, N. N. Xu, H. E. D. J. Liu, and C. L. Wang, A first-principles study on magnetic properties of the intrinsic defects in wurtzite ZnO, *J. Chem. Phys.* **150**, 094704 (2019).
- [81] F. Tuomisto, V. Ranki, K. Saarinen, and D. C. Look, Evidence of the Zn vacancy acting as the dominant acceptor in n-type ZnO, *Phys. Rev. Lett.* **91**, 205502 (2003).
- [82] D. Savchenko, A. Vasin, O. Kuz, I. Verovsky, A. Prokhorov, A. Nazarov, J. Lančok, and E. Kalabukhova, Role of the paramagnetic donor-like defects in the high n-type conductivity of the hydrogenated ZnO microparticles, *Sci. Rep.* **10**, 17347 (2020).
- [83] J. R. Maze, A. Gali, E. Togan, Y. Chu, A. Trifonov, E. Kaxiras, and M. D. Lukin, Group theoretical analysis of nitrogen-vacancy center's energy levels and selection rules, *MRS Proc.* **1282**, mrsf10 (2011).
- [84] T. J. Smart, K. Li, J. Xu, and Y. Ping, Intersystem crossing and exciton-defect coupling of spin defects in hexagonal boron nitride, *npj Comput. Mater.* **7**, 1 (2021).
- [85] M. J. Rayson, and P. R. Briddon, First principles method for the calculation of zero-field splitting tensors in periodic systems, *Phys. Rev. B* **77**, 035119 (2008).
- [86] F. Neese, Calculation of the zero-field splitting tensor on the basis of hybrid density functional and Hartree-Fock theory, *J. Chem. Phys.* **127**, 164112 (2007).
- [87] R. E. Coffman, M. I. Himaya, and K. Nyeu, Spin-Hamiltonian parameters and spin-orbit coupling for V^{3+} in ZnO, *Phys. Rev. B* **4**, 3250 (1971).
- [88] G. Filipovich, A. L. Taylor, and R. E. Coffman, Electron paramagnetic resonance of V^{3+} ions in zinc oxide, *Phys. Rev. B* **1**, 1986 (1970).
- [89] V. Ivády, I. A. Abrikosov, and A. Gali, First principles calculation of spin-related quantities for point defect qubit research, *npj Comput. Mater.* **4**, 1 (2018).
- [90] A. Lenef, and S. C. Rand, *Phys. Rev. B* **53**, 13441 (1996).
- [91] M. W. Doherty, N. B. Manson, P. Delaney, and L. C. L. Hollenberg, The negatively charged nitrogen-vacancy centre in diamond: The electronic solution, *New J. Phys.* **13**, 025019 (2011).
- [92] S. V. Streltsov, and D. I. Khomskii, Jahn-Teller effect and spin-orbit coupling: Friends or Foes?, *Phys. Rev. X* **10**, 031043 (2020).
- [93] I. Bersuker, *The Jahn-Teller Effect* (Cambridge University Press, Cambridge, 2006).
- [94] L. Razinkovas, M. W. Doherty, N. B. Manson, C. G. Van de Walle, and A. Alkauskas, Vibrational and vibronic structure of isolated point defects: The nitrogen-vacancy center in diamond, *Phys. Rev. B* **104**, 045303 (2021).
- [95] I. B. Bersuker, The Jahn-Teller and pseudo Jahn-Teller effect in materials science, *J. Phys.: Conf. Ser.* **833**, 012001 (2017).
- [96] G. Thiering, and A. Gali, Theory of the optical spin-polarization loop of the nitrogen-vacancy center in diamond, *Phys. Rev. B* **98**, 085207 (2018).
- [97] G. Thiering, and A. Gali, Ab initio magneto-optical spectrum of group-IV vacancy color centers in diamond, *Phys. Rev. X* **8**, 021063 (2018).
- [98] J. Heyd, G. E. Scuseria, and M. Ernzerhof, Hybrid functionals based on a screened Coulomb potential, *J. Chem. Phys.* **118**, 8207 (2003).

- [99] G. Kresse, and J. Furthmüller, Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [100] G. Kresse, and J. Furthmüller, Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set, *Comput. Mater. Sci.* **6**, 15 (1996).
- [101] G. Kresse, and J. Hafner, Ab initio molecular-dynamics simulation of the liquid-metal-amorphous-semiconductor transition in germanium, *Phys. Rev. B* **49**, 14251 (1994).
- [102] M. Thomas, Semiconductors—Basic data, 2nd ed., edited by O. Madelung (Springer, Berlin, 1996, viii, 317 pp., hardcover, DM 88.00, ISBN 3-540-60883-4, *Chem. Vapor Deposition* **3**, 288 (1997).
- [103] J. Albertsson, S. C. Abrahams, and A. Kvik, Atomic displacement, anharmonic thermal vibration, expansivity and pyroelectric coefficient thermal dependences in ZnO, *Acta Crystallogr. Sect. B* **45**, 34 (1989).
- [104] F. Oba, A. Togo, I. Tanaka, J. Paier, and G. Kresse, Defect energetics in ZnO: A hybrid Hartree-Fock density functional study, *Phys. Rev. B* **77**, 245202 (2008).
- [105] D. C. Reynolds, D. C. Look, B. Jogai, C. W. Litton, G. Cantwell, and W. C. Harsch, Valence-band ordering in ZnO, *Phys. Rev. B* **60**, 2340 (1999).
- [106] J. Ning, J. W. Furness, and J. Sun, Reliable lattice dynamics from an efficient density functional approximation, *Chem. Mater.* **34**, 2562 (2022).
- [107] R. Sundararaman, and Y. Ping, First-principles electrostatic potentials for reliable alignment at interfaces and defects, *J. Chem. Phys.* **146**, 104109 (2017).
- [108] F. Wu, A. Galatas, R. Sundararaman, D. Rocca, and Y. Ping, First-principles engineering of charged defects for two-dimensional quantum technologies, *Phys. Rev. Mater.* **1**, 071001(R) (2017).
- [109] F. Oba, and Y. Kumagai, Design and exploration of semiconductors from first principles: A review of recent advances, *Appl. Phys. Express* **11**, 060101 (2018).
- [110] D. Sangalli, A. Ferretti, H. Miranda, C. Attaccalite, I. Marri, E. Cannuccia, P. Melo, M. Marsili, F. Paleari, A. Marrazzo, G. Prandini, P. Bonfà, M. O. Atambo, F. Affinito, M. Palummo, A. Molina-Sánchez, C. Hogan, M. Grüning, D. Varsano, and A. Marini, Many-body perturbation theory calculations using the yambo code, *J. Phys.: Condens. Matter* **31**, 325902 (2019).
- [111] P. Giannozzi *et al.*, QUANTUM ESPRESSO: A modular and open-source software project for quantum simulations of materials, *J. Phys.: Condens. Matter* **21**, 395502 (2009).
- [112] D. R. Hamann, Optimized norm-conserving Vanderbilt pseudopotentials, *Phys. Rev. B* **88**, 085117 (2013).
- [113] M. Schlupf, and F. Gygi, Optimization algorithm for the generation of ONCV pseudopotentials, *Comput. Phys. Commun.* **196**, 36 (2015).
- [114] F. Wu, D. Rocca, and Y. Ping, Dimensionality and anisotropy dependence of radiative recombination in nanostructured phosphorene, *J. Mater. Chem. C* **7**, 12891 (2019).
- [115] K. Czelej, M. R. Zemła, P. Śpiewak, and K. J. Kurzydowski, Quantum behavior of hydrogen-vacancy complexes in diamond, *Phys. Rev. B* **98**, 235111 (2018).
- [116] K. Czelej, M. R. Lambert, M. E. Turiansky, A. Koshevnikov, S. Mu, and C. G. Van de Walle, Transition-metal-related quantum emitters in wurtzite AlN and GaN, *ACS Nano* **18**, 28724 (2024).
- [117] A. Gali, M. Fyta, and E. Kaxiras, Ab initio supercell calculations on nitrogen-vacancy center in diamond: Electronic structure and hyperfine tensors, *Phys. Rev. B* **77**, 155206 (2008).
- [118] L. Shang, Q. Chen, W. Jing, C.-G. Ma, C.-K. Duan, and J. Du, First-principles study of transition metal dopants as spin qubits, *Phys. Rev. Mater.* **6**, 086201 (2022).
- [119] A. V. Ivanov, Y. L. A. Schmerwitz, G. Levi, and H. Jónsson, Electronic excitations of the charged nitrogen-vacancy center in diamond obtained using time-independent variational density functional calculations, *SciPost Phys.* **15**, 009 (2023).
- [120] M. D. McCluskey, in *Semiconductors and Semimetals*, Defects in Semiconductors, Vol. 91, edited by L. Romano, V. Privitera, and C. Jagadish (Elsevier, 2015), pp. 279–313.
- [121] H. Seo, A. L. Falk, P. V. Klimov, K. C. Miao, G. Galli, and D. D. Awschalom, Quantum decoherence dynamics of divacancy spins in silicon carbide, *Nat. Commun.* **7**, 12935 (2016).
- [122] H. Park, J. Lee, S. Han, S. Oh, and H. Seo, Decoherence of nitrogen-vacancy spin ensembles in a nitrogen electron-nuclear spin bath in diamond, *npj Quantum Inf.* **8**, 1 (2022).
- [123] J. Lee, H. Park, and H. Seo, First-principles theory of extending the spin qubit coherence time in hexagonal boron nitride, *npj 2D Mater. Appl.* **6**, 1 (2022).
- [124] W. Yang, and R.-B. Liu, Quantum many-body theory of qubit decoherence in a finite-size spin bath, *Phys. Rev. B* **78**, 085315 (2008).
- [125] F. Neese, The ORCA program system, *Wiley Interdiscip. Rev. Comput. Mol. Sci.* **2**, 73 (2012).
- [126] F. Neese, F. Wennmohs, U. Becker, and C. Riplinger, The ORCA quantum chemistry program package, *J. Chem. Phys.* **152**, 224108 (2020).
- [127] F. Weigend, and R. Ahlrichs, Balanced basis sets of split valence, triple zeta valence and quadruple zeta valence quality for H to Rn: Design and assessment of accuracy, *Phys. Chem. Chem. Phys.* **7**, 3297 (2005).
- [128] M. Reiher, Douglas-Kroll-Hess theory: A relativistic electrons-only theory for chemistry, *Theor. Chem. Acc.* **116**, 241 (2006).
- [129] D. A. Pantazis, X.-Y. Chen, C. R. Landis, and F. Neese, All-electron scalar relativistic basis sets for third-row transition metal atoms, *J. Chem. Theory Comput.* **4**, 908 (2008).
- [130] J. D. Rolfes, F. Neese, and D. A. Pantazis, All-electron scalar relativistic basis sets for the elements Rb–Xe, *J. Comput. Chem.* **41**, 1842 (2020).
- [131] F. Neese, Efficient and accurate approximations to the molecular spin-orbit coupling operator and their use in molecular *g*-tensor calculations, *J. Chem. Phys.* **122**, 034107 (2005).

- [132] X. Huang, E. Lindgren, and J. R. Chelikowsky, Surface passivation method for semiconductor nanostructures, *Phys. Rev. B* **71**, 165328 (2005).
- [133] E. Badaeva, Y. Feng, D. R. Gamelin, and X. Li, Investigation of pure and Co^{2+} -doped ZnO quantum dot electronic structures using the density functional theory: Choosing the right functional, *New J. Phys.* **10**, 055013 (2008).
- [134] Input files, post-processing scripts, and source data for this paper are available in https://github.com/Ping-Group-UCSC/ZnO_project_data (2026).
- [135] T. J. Boerner, S. Deems, T. R. Furlani, S. L. Knuth, and J. Towns, in *Practice and Experience in Advanced Research Computing 2023: Computing for the Common Good*, PEARC '23 (Association for Computing Machinery, New York, NY, USA, 2023), pp. 173–176.