

— Supporting Information —

Strategies to search for two-dimensional materials with long spin qubit coherence time

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Supplementary Note 1: List of substrates

Table S1: List of substrates tested in this study.

Substrate	Miller indices
Diamond	(0 0 1), (1 1 1)
Graphite	(0 0 1)
Si	(0 0 1), (1 1 1)
SiC (3C)	(0 0 1)
SiO ₂ (Quartz)	(0 0 1), (0 1 0), (1 0 0)
CaO	(0 0 1), (0 1 1), (1 1 1)
CeO ₂	(0 0 1), (0 1 1), (1 1 1)

Supplementary Note 2: Coherence and chemical composition

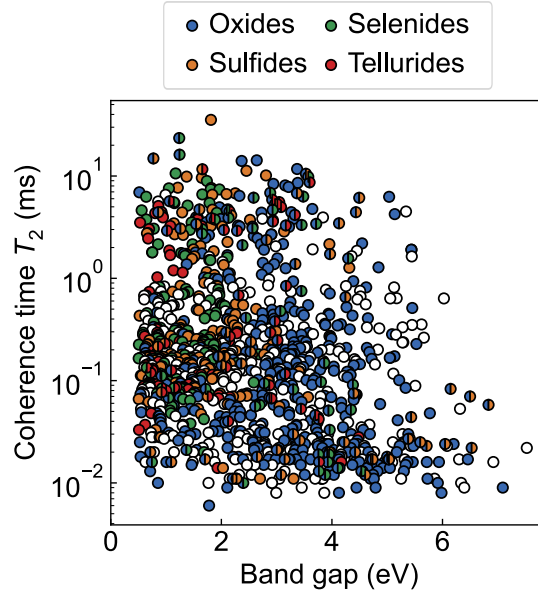


Figure S1: Coherence times (T_2) of 2D materials computed using the cluster correlation expansion (CCE) approach. The band gaps of these monolayers are taken from the MC2D database, which tabulates values calculated using density functional theory with the Perdew-Burke-Ernzerhof (PBE) functional. The colors represent the chalcogen chemistry of each material: white points represent monolayers with no chalcogen atoms, and multi-colored points represent monolayers with multiple chalcogens in its chemical makeup. The vast chemical diversity of 2D materials with high T_2 can provide flexibility in designing quantum devices with specific functions.

Supplementary Note 3: Strain-induced change in T_2

In our study, we create lattice-matched heterostructures between 2D materials and 3D substrates, ensuring that the strain applied to the 2D material is no more than 5%. Here, we calculate the effect of applying strain on T_2 of the bare 2D host, which is important when considering the origin of the substrate-induced decrease of T_2 in heterostructural qubit hosts. By comparing the T_2 of the unstrained 2D host against that of the strained host, we find that T_2 of the bare 2D host varies by no more than 20% when the applied strain is less than 5% (Figure S2). This indicates that substrate-induced strain cannot explain the substantial decrease in T_2 from the bare 2D host material to a heterostructural host system, which can be orders of magnitude when the substrate is, e.g., SiO_2 .

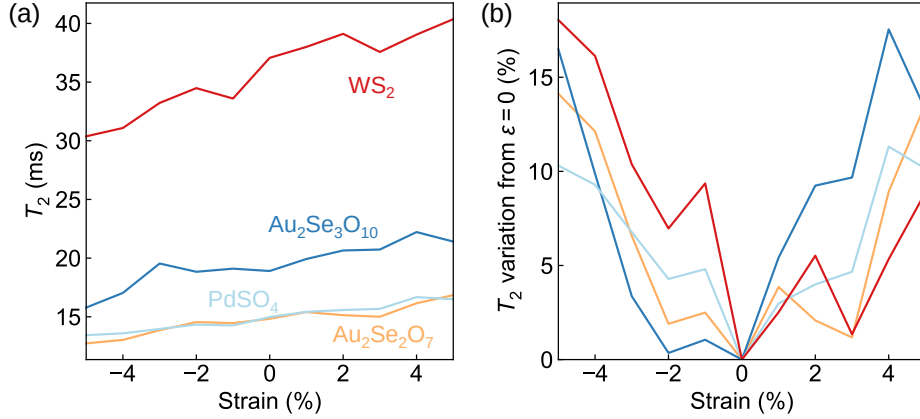


Figure S2: (a) Coherence times (T_2) of biaxially-strained 2D materials computed using the cluster correlation expansion (CCE) approach, considering only hyperfine interactions. (b) Variation in T_2 of the strained 2D host compared to that of the unstrained material. The variation is computed using the formula $|T_2(\varepsilon) - T_2(\varepsilon = 0)|/T_2(\varepsilon = 0)$.

Supplementary Note 4: Effects of interfacial reconstruction on the predicted T_2 in heterostructures

Since we do not relax the interface in our lattice-matched heterostructures with e.g. *ab initio* methods, it is natural to ask whether specific details of the interface affect the predicted nuclear spin bath-driven T_2 of heterostructures. To address this question, we perform two checks.

Firstly, there can be variations in T_2 due to the interlayer separation distance (d) between the 2D host and substrate. In our study, we approximate d as the sum of van der Waals radii of atoms at the interface (i.e., $d \approx d_{\text{vdW}}$). However, uncertainties in d can arise from interfacial reconstruction effects and even the tabulated van der Waals radii themselves.^{1,2} We therefore test the variation of T_2 as a function of d from $0.8d_{\text{vdW}}$ to $1.2d_{\text{vdW}}$. The variation (i.e. error) in T_2 is calculated using the formula

$$\text{Error} = \frac{|T_2(d = d_{\text{vdW}}) - T_2(d)|}{T_2(d)} \times 100. \quad (\text{S1})$$

In other words, we calculate the error in T_2 by assuming $d = d_{\text{vdW}}$ as the interlayer separation distance, if $d \neq d_{\text{vdW}}$ in the experimental (i.e. true) heterostructure. Our results show that such an error is no more than 10% (Figure S3), suggesting that T_2 is relatively insensitive to variations in d .

Secondly, atomic reconstruction near the interface can potentially impact coherence properties. We test the impact of surface reconstruction on T_2 by considering models of the heterostructures where atoms in the substrate are randomly displaced, but the atoms near the interface are allowed to be displaced more than those far from the interface. In particular, we let each atom displace in a random direction, where the magnitude of the displacement is determined by $w = 0.5 \exp(-10z)$ Å, where z is the relative depth of the atom from the interface ($z = 0$ at the interface, $z = 1$ at the bottom of the substrate). In other words, we consider an extreme case of interfacial reconstruction, where atoms near the interface are essentially amorphized. Despite this structural change, out of 10 such structures for each heterostructural combination, we find that T_2 does not stray from that of the ideal heterostructure (where the atoms are not displaced) by more than 15% (Figure S4).

Our results here strongly suggest that T_2 is more dependent on the nuclear spin environment of the substrate material, rather than the interfacial geometry. We note, however, that we have not tested the influence of e.g. electric noise caused by dangling bonds or defects formed at the heterointerface.

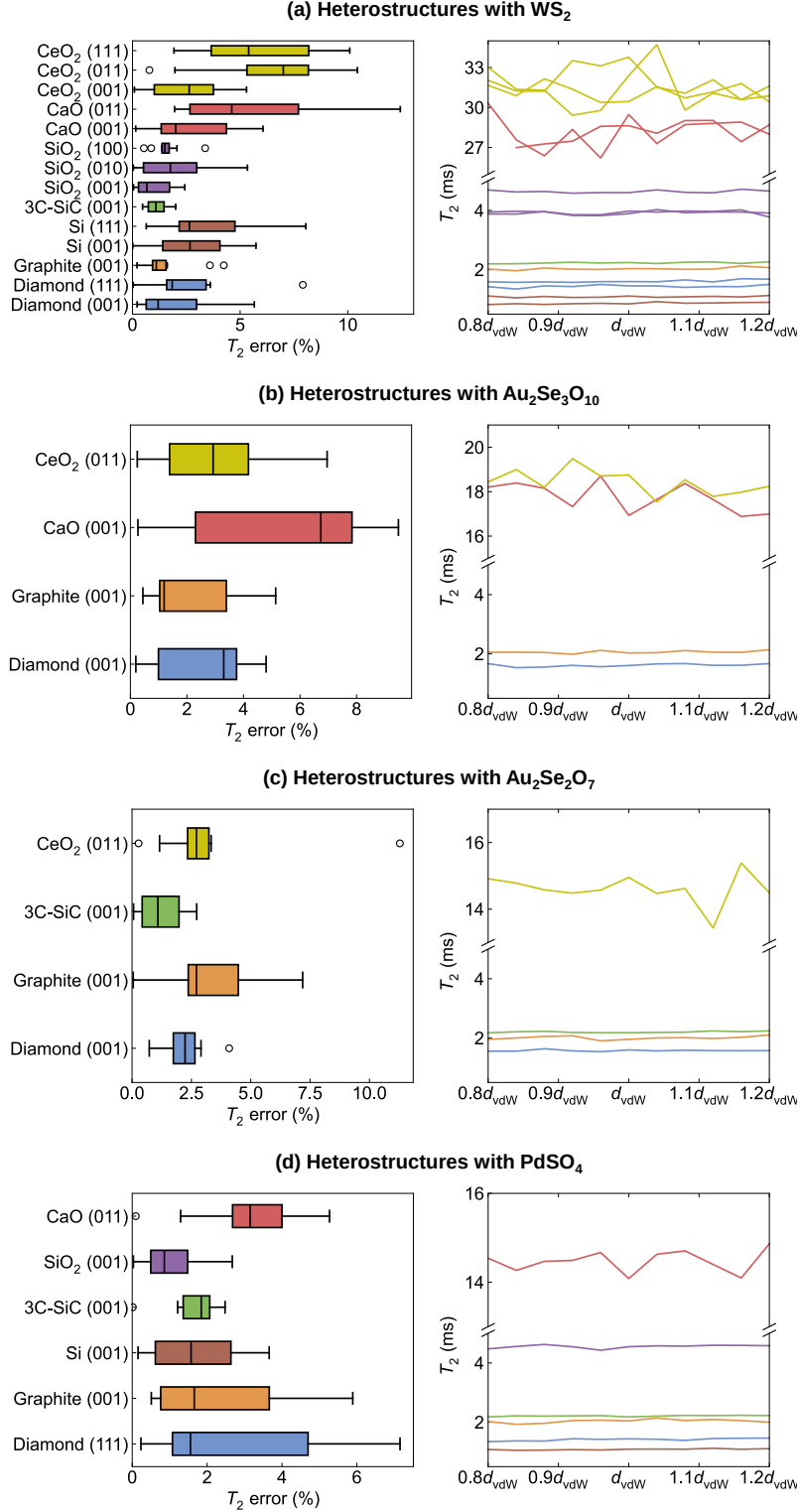


Figure S3: Coherence times (T_2) of heterostructures where the 2D host material is (a) WS₂, (b) Au₂Se₃O₁₀, (c) Au₂Se₂O₇, and (d) PdSO₄. The colors represent the substrate. T_2 is calculated by varying the interlayer separation between the 2D material and substrate, from $d = 0.8d_{vdW}$ to $d = 1.2d_{vdW}$, where d_{vdW} is the sum of the van der Waals radii of the atoms at the interface. The calculated T_2 as a function of d are shown in the right panels, and the errors (in %) relative to the value of T_2 computed at $d = d_{vdW}$ are shown in the left panels.

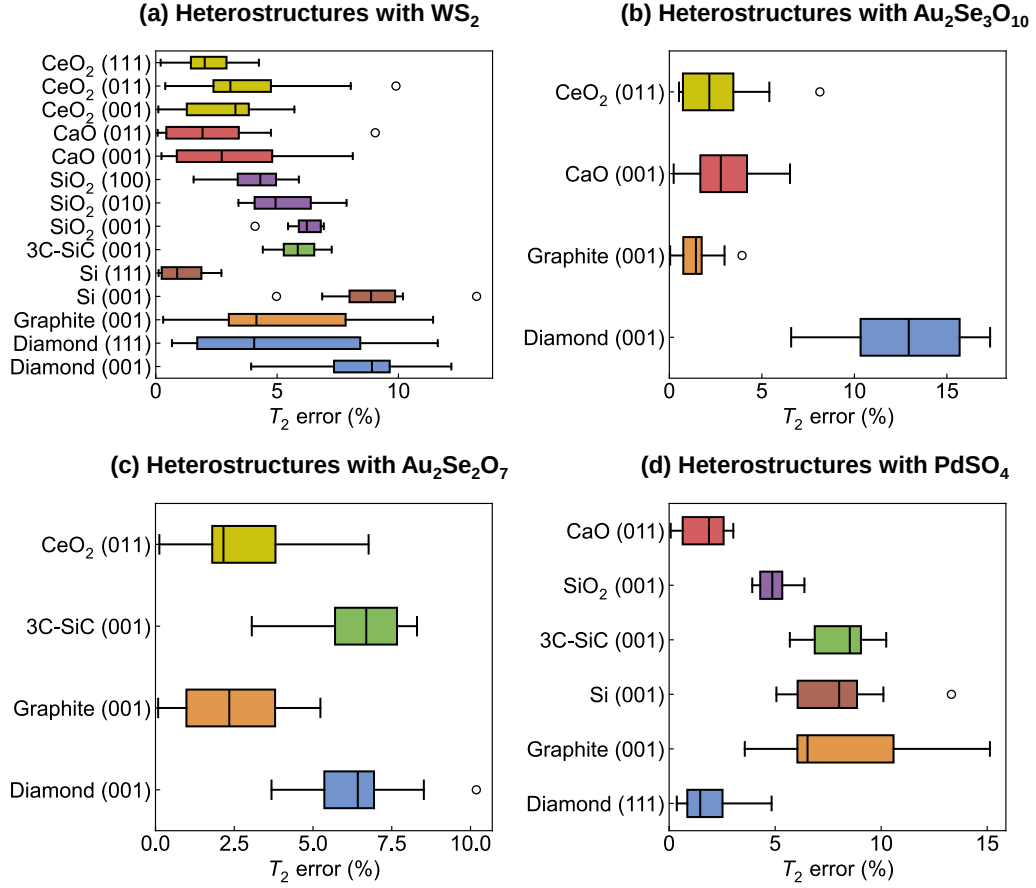


Figure S4: The error in coherence times (T_2) between a perfect (unrelaxed) heterostructure and models of relaxed heterostructures. The errors are shown for heterostructures where the 2D host material is (a) WS_2 , (b) $\text{Au}_2\text{Se}_3\text{O}_{10}$, (c) $\text{Au}_2\text{Se}_2\text{O}_7$, and (d) PdSO_4 . The colors represent the substrate material. For each heterostructural combination, T_2 is calculated on 10 different structures, which are generated by randomly displacing atoms in the substrate material.

Supplementary Note 5: Heteronuclear decoupling in 2D materials

It is important to check whether heteronuclear spin baths can be decoupled in 2D host materials, similar to how they are often decoupled in 3D systems.^{3,4} Within the CCE approach, the coherence function is factorized into interactions between the spin qubit and single nuclear spins, pairs of nuclear spins, and so forth. Assuming CCE-2 level of theory, the coherence function can be written as

$$\mathcal{L}(t) = \prod_i \tilde{\mathcal{L}}_i(t) \prod_{\{i,j\}} \tilde{\mathcal{L}}_{i,j}(t) \quad (\text{S2})$$

where i and j represent nuclear spins, and $\tilde{\mathcal{L}}_i(t)$ and $\tilde{\mathcal{L}}_{i,j}$ are contributions to the coherence decay from interactions with single nuclear spins and pairs of nuclear spins, respectively. Pairs of nuclear spins can be made between the same (homonuclear) or different (heteronuclear) isotopes. Accordingly, one can write

$$\prod_{\{i,j\}} \tilde{\mathcal{L}}_{i,j}(t) = \underbrace{\prod_i \tilde{\mathcal{L}}_{i,i}(t)}_{\text{homonuclear}} \underbrace{\prod_{i \neq j} \tilde{\mathcal{L}}_{i,j}(t)}_{\text{heteronuclear}} \quad (\text{S3})$$

and

$$\mathcal{L}(t) = \prod_i \tilde{\mathcal{L}}_i(t) \prod_i \tilde{\mathcal{L}}_{i,i}(t) \prod_{i \neq j} \tilde{\mathcal{L}}_{i,j}(t) \quad (\text{S4})$$

$$\equiv \prod_n \tilde{\mathcal{L}}_{\text{homo}_n}(t) \prod_{i \neq j} \tilde{\mathcal{L}}_{i,j}(t) \quad (\text{S5})$$

where n is the type of nuclear isotope (e.g. ^{183}W and ^{33}S in WS_2), distinct from the individual isotopes i and j in the nuclear spin bath.

The contribution from the heteronuclear pairs in each material can be understood by dividing each homonuclear spin bath $\tilde{\mathcal{L}}_{\text{homo}_n}(t)$ from the total coherence decay profile $\mathcal{L}(t)$. We find that the contribution from heteronuclear pairs is nearly 1 within the simulated time domain (Figure S5), which indicates that heteronuclear spin baths can be decoupled into contributions from homonuclear spin baths in 2D materials, i.e.

$$\mathcal{L}(t) \approx \prod_n \tilde{\mathcal{L}}_{\text{homo}_n}(t). \quad (\text{S6})$$

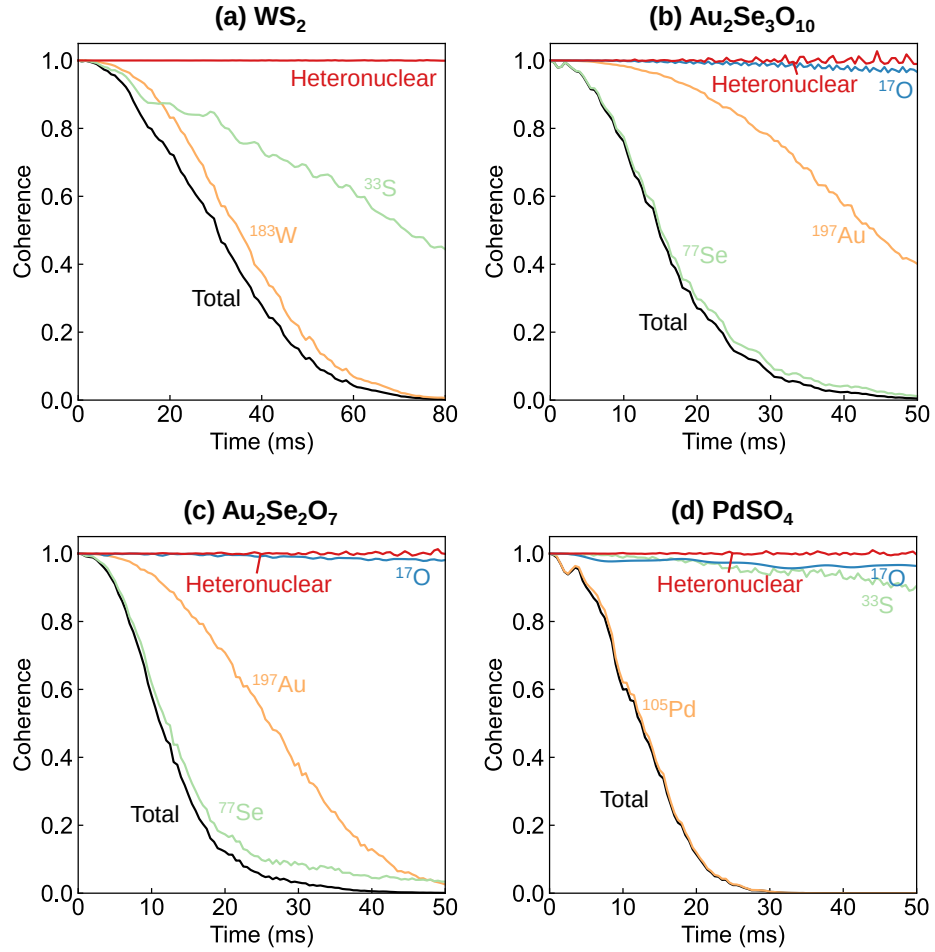


Figure S5: Coherence functions of (a) WS_2 , (b) $\text{Au}_2\text{Se}_3\text{O}_{10}$, (c) $\text{Au}_2\text{Se}_2\text{O}_7$, and (d) PdSO_4 , shown alongside contributions from each homonuclear spin bath (labeled by the nuclear isotope) and the heteronuclear spin bath. The calculations are performed at a magnetic field strength of 300 mT.

Supplementary Note 6: T_2 model for 2D materials

Derivation and fitting

Kanai et al. proposed a model of T_2 for a spin qubit in a host material, based on scaling laws of T_2 with isotopic features.⁴ Within the model, the coherence time is calculated as

$$T_{2,\text{Kanai}}^{-\eta} = \sum_i T_{2,\text{Kanai},i}^{-\eta} \quad (\text{S7})$$

where the stretch exponent η is assumed to be 2. The sum is performed over all isotopes i with nonzero nuclear spin in the host material, and

$$T_{2,\text{Kanai},i} = 1.5 \times 10^{18} |g_i|^{-1.6} I_i^{-1.1} n_i^{-1.0} (s) \quad (\text{S8})$$

is the coherence time for a particular isotope i , where g_i is the g -factor, I_i is the nuclear spin, and n_i is the density of nuclear spins.

Davis et al.⁵ proposed a model for the coherence function of an interacting many-body system described by the Hamiltonian

$$\hat{H} = \sum_i \frac{J_z}{r_i^\alpha} \hat{S}_q^z \hat{S}_i^z \quad (\text{S9})$$

where J_z is a factor representing the coupling strength, $\hat{S}_q^z$ is the qubit spin, and $\hat{S}_i^z$ is the spin of the i th nuclear spin. The interaction is assumed to follow the power-law form $1/r^\alpha$. From the Hamiltonian $\hat{H}$, the coherence function of a qubit can be derived:⁵

$$\mathcal{L}(t) = \exp \left(-n A_D \frac{D}{\alpha} \left[-\frac{\Gamma(-\frac{D}{2\alpha})}{2^{\frac{D}{2\alpha}+1}} \right] \left[\frac{\bar{g} J_z}{2} \right]^{\frac{D}{\alpha}} \chi(t)^{\frac{D}{2\alpha}} \right), \quad (\text{S10})$$

where

$$\chi(t) = \frac{t^3}{6\tau_c} \quad (\text{S11})$$

is the response function for a Hahn-echo pulse sequence in the early-time regime. n is the concentration of nuclear isotopes, A_D is the (dimensionless) unit volume of a D -dimensional sphere, $\Gamma(x)$ is the Γ -function, $\bar{g}$ accounts for the angular dependence of the interaction, and τ_c is the correlation time.

If we fit $\mathcal{L}(t)$ to a stretched exponential function

$$\mathcal{L}(t) \equiv \exp \left(-\left(\frac{t}{T_{2,D}} \right)^{\eta_D} \right), \quad (\text{S12})$$

then we have

$$\eta_D = \frac{3D}{2\alpha} \quad (\text{S13})$$

and

$$\begin{aligned} T_{2,D} &= \left(nA_D \frac{D}{\alpha} \left[-\frac{\Gamma(-\frac{D}{2\alpha})}{2^{\frac{D}{2\alpha}+1}} \right] \left[\frac{\bar{g}J_z}{2} \right]^{\frac{D}{\alpha}} \left(\frac{1}{6\tau_c} \right)^{\frac{D}{2\alpha}} \right)^{-1/\eta_D} \\ &= \left(nA_D \frac{D}{\alpha} \left[-\frac{\Gamma(-\frac{D}{2\alpha})}{2^{\frac{D}{2\alpha}+1}} \right] \right)^{-1/\eta_D} \left[\frac{\bar{g}J_z}{2} \right]^{-\frac{2}{3}} \left(\frac{1}{6\tau_c} \right)^{-\frac{1}{3}}. \end{aligned} \quad (\text{S14})$$

Note that the units are matched in the expression, because n is the nuclear spin density with units of (distance) $^{-D}$, J_z is the coupling parameter with units of (distance) $^\alpha$ /(time), τ_c has units of time, and all other parameters are dimensionless.

Using Eq. (S14), we can find a dimension-dependent scaling factor for T_2 . In particular, if we have a prediction for T_2 for a 3D material, then we can derive a scaling law that enables predictions of T_2 for any D -dimensional version of the material. Here, we are interested in building upon the model proposed by Kanai et al. for 3D systems as shown in Eqs. (S17) and (S8).⁴ Because heteronuclear spin baths can be decoupled in a 2D nuclear spin environment (Supplementary Note 5), we apply the dimension-dependent scaling to the contribution from each individual nuclear isotope,

$$\frac{T_{2,2D,i}}{T_{2,3D,i}} = \frac{T_{2,2D,i}}{T_{2,\text{Kanai},i}} = C(\alpha_{2D}) \frac{n_i^{2/3}}{(n_i w)^{\alpha_{2D}/3}}, \quad (\text{S15})$$

where

$$C(\alpha_{2D}) \equiv \frac{\left(A_3 \left[-\frac{\Gamma(-\frac{1}{2})}{2^{\frac{3}{2}}} \right] \right)^{2/3}}{\left(\frac{2}{\alpha_{2D}} A_2 \left[-\frac{\Gamma(-\frac{1}{\alpha_{2D}})}{2^{\frac{1}{\alpha_{2D}}+1}} \right] \right)^{\alpha_{2D}/3}} \quad (\text{S16})$$

is a constant with units (distance) $^{2-(2\alpha_{2D}/3)}$, and we consider the density n_i of the specific isotope i , and w is the thickness of the monolayer. The overall T_2 for the 2D host material can then be calculated as

$$T_{2,2D}^{-\eta_2} = \sum_i T_{2,2D,i}^{-\eta_2} = \sum_i T_{2,2D,i}^{-3/\alpha_{2D}}, \quad (\text{S17})$$

where we invoke Eq. (S13) in the second equivalence.

We note some assumptions in our derivation. We assume that the numerical values of $\bar{g}$, J_z , and τ_c get canceled in Eq. (S16). In other words, we assume that the nuclear spin bath is homogeneously-distributed

around the qubit, thus leading to an effectively isotropic interaction with the nuclear spins ($\bar{g} \approx 1$), and that the coupling strength prefactor does not change across dimensions as it depends on properties (e.g. the g-factor) which are independent of dimensionality. Additionally, τ_c depends on the driving frequency of the experimental setup.⁵ Since τ_c has been shown to be within the same order of magnitude in 2D and 3D environments of P1 centers in diamond when a consistent driving frequency is applied,⁵ we assume that $\tau_{c,2D} \approx \tau_{c,3D}$ in our derivation. We also let $\alpha_{3D} = 3$ for dipolar interactions in 3D from classical electrostatics, which naturally implies $\eta_3 = 1.5$. We discuss these assumptions in further detail in the following subsection entitled “Considerations”.

There is one undetermined parameter, α_{2D} , representing the power-law interaction between the spin qubit and nuclear spins in a 2D material (see Eq. S9). We determine the best-fit α_{2D} by minimizing the mean relative error between the model T_2 and CCE-calculated T_2 , defined by

$$\text{Error}(\alpha_{2D}) = \frac{1}{N} \sum_{x \in \{\text{data}\}} \frac{|T_{2,2D,x}(\alpha_{2D}) - T_{2,\text{CCE},x}|}{T_{2,\text{CCE},x}}. \quad (\text{S18})$$

where N is the number of CCE data points. In other words, we determine

$$\alpha_{2D,\text{fitted}} = \underset{\alpha_{2D}}{\text{argmin}} [\text{Error}(\alpha_{2D})]. \quad (\text{S19})$$

We find that $\alpha_{2D} = 2.84$ yields the best fit. As a result, the contribution from a single nuclear isotope i in a 2D environment is

$$T_{2,2D,i} \approx 0.94w^{-0.95}n_i^{-0.28}T_{2,\text{Kanai},i}, \quad (\text{S20})$$

and the overall T_2 for a qubit in a 2D host can be predicted using the formula

$$T_{2,2D}^{-1.06} \approx \sum_i T_{2,2D,i}^{-1.06}. \quad (\text{S21})$$

Considerations

It is important to comment on the stretch exponent for 3D systems, which is set to $\eta_3 = 1.5$ in this study. This is justified by the use of Eq. (S13) and the fact that dipolar interactions in 3D are described by a $\sim 1/r^3$ power-law form (i.e. $\alpha_{3D} = 3$).⁶ Our fitting implies that dipolar interactions in 2D can be described, on average, by an effective $\sim 1/r^{2.84}$ power-law form, which is slightly longer-ranged than in 3D. This is physically consistent with the reduced dielectric screening in a 2D spin environment.⁶

In the study by Kanai et al., the authors assumed a stretch exponent of $\eta_3 = 2$ for 3D systems. If we

set $\eta_3 = 2$ and re-fit our model for 2D systems, we obtain $\alpha_{2D} = 2.12$. While the accuracies of the models are comparable (Table S2), setting the 3D stretch exponent to $\eta_3 = 1.5$ enables a physically-justifiable interpretation of the fitted α_{2D} value within the semi-classical framework of Ref. 5.

Table S2: Comparison of fitted α_{2D} and mean relative error for different values of the 3D stretch exponent η_3 . The mean relative error is calculated using Eq. (S18).

η_3	α_{3D}	α_{2D} (fitted)	Error
1.5	3	2.84	0.38
2	2.25	2.12	0.35

The mean relative error can potentially be reduced in several ways, for example by using machine learning tools. Within our theoretical framework, one may consider fitting an additional undetermined prefactor term in Eq. (S16) due to uncertainties in how parameters such as τ_c vary across dimensions, i.e.

$$C(\alpha_{2D}) \rightarrow A_{\text{prefactor}} C(\alpha_{2D}). \quad (\text{S22})$$

However, we find that including such a prefactor term and re-running the analysis with $\eta_3 = 1.5$ results in a mean relative error of 0.33 which, compared to our fitting procedure without the prefactor term (MRE = 0.38), does not improve the model considerably. Moreover, including the undetermined prefactor term in our analysis results in an unjustifiable fitted value of $\alpha_{2D} = 2.20$, which is significantly lower than the value for hyperfine interactions in 3D ($\alpha_{3D} = 3$).

Implementation

We apply the T_2 model (Eqs. S20 and S21) to 2D materials curated by the following databases: 2DMatPedia,⁷ C2DB,^{8,9} and an unnamed database created using symmetry considerations¹⁰ (which we refer to as the “symmetry-based database”). We initially screen these databases for materials with GGA band gap greater than 0.5 eV, using the same threshold as in our procedure with the MC2D database. We then check for stability of the material by using the information available in each respective database. For the C2DB and symmetry-based databases, we screen for materials that have convex hull energy less than 0.2 eV/atom. For 2DMatPedia, we retain materials with decomposition energy less than 0.1 eV/atom and exfoliation energy less than 0.2 eV/atom. The decomposition energy is defined as the energy required to decompose the 2D material into a more stable phase with the same chemical composition, excluding the 3D layered parent phase from which the 2D material originates.⁷ The stability thresholds are determined by the fact that most experimentally-known 2D materials have a calculated hull energy (for the C2DB and symmetry-based

databases) and calculated decomposition energy (for 2DMatPedia) within the threshold value.^{7,8} We lastly remove any duplicate structures by employing a structure matching algorithm, where information about the local coordination of each site in a structure are encoded into features and compared with the features of other structures.¹¹ The T_2 model (Eqs. S20 and S21) is then applied to the remaining structures. We disregard any material that contains an element with no stable isotope (e.g., Tc).

Supplementary Note 7: T_2 model for heterostructures

Derivation and fitting

We consider a model where the spin qubit is implanted in a 2D material which stretches infinitely in the x and y directions. The 2D material is placed on a 3D substrate material, which also stretches infinitely in the x and y directions, as well as in the $-z$ direction below the 2D material (Figure S6).

From the many-body Hamiltonian in Eq. (S9), we can express the coherence function $\mathcal{L}(t)$ as⁵

$$\begin{aligned}\mathcal{L}(t) &= \exp \left(-n \int \left[1 - e^{-\frac{1}{2} \left(\frac{J_z |g| \chi(t)^{1/2}}{2r^\alpha} \right)^2} \right] d\mathbf{r} \right) \\ &\equiv \exp \left(-n \int F(r) d\mathbf{r} \right).\end{aligned}\tag{S23}$$

We decouple a heteronuclear spin bath into homonuclear baths of individual isotopes, as explained in [Supplementary Note 5](#). In other words,

$$\begin{aligned}\mathcal{L}(t) &= \prod_i \mathcal{L}_i(t) \\ &= \exp \left(\sum_i -n_i \int F_i(r) d\mathbf{r} \right),\end{aligned}\tag{S24}$$

where i represents each isotope type in the heterostructure. In other words, the summation represents all contributions from each individual homonuclear spin bath. The set of all nuclear isotopes in the heterostructure can be categorized into three subsets (in set notation):

1. The subset of isotopes in the 2D host material but not in the substrate, $\{\text{host}\} \setminus \{\text{sub.}\}$,

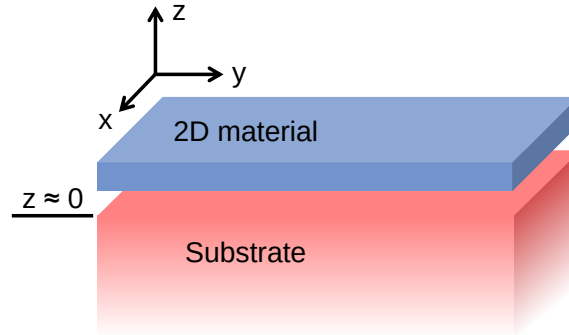


Figure S6: Structural model considered when deriving T_2 for heterostructures.

2. The subset of isotopes in the substrate but not in the 2D host material, $\{\text{sub.}\} \setminus \{\text{host}\}$, and
3. The subset of isotopes which exist in both the 2D host material and the substrate, $\{\text{host}\} \cap \{\text{sub.}\}$.

Accordingly, $\mathcal{L}(t)$ can be re-expressed as

$$\mathcal{L}(t) = \exp \left(\left[\sum_{i \in \{\text{host}\} \setminus \{\text{sub.}\}} -n_i \int_{\text{host}} F_i(r) d\mathbf{r} \right] + \left[\sum_{j \in \{\text{sub.}\} \setminus \{\text{host}\}} -n_j \int_{\text{sub.}} F_j(r) d\mathbf{r} \right] + \left[\sum_{k \in \{\text{host}\} \cap \{\text{sub.}\}} -n_k \int F_k(r) d\mathbf{r} \right] \right). \quad (\text{S25})$$

As an example, in the $\text{Au}_2\text{Se}_3\text{O}_{10}/\text{CaO}$ heterostructure, we have $i \in \{^{197}\text{Au}, ^{77}\text{Se}\}$, $j \in \{^{43}\text{Ca}\}$, and $k \in \{^{17}\text{O}\}$.

The last summand in the exponent in Eq. (S25) accounts for isotopes k in both the 2D host material and substrate material. The domain of the integration, which accounts for the positional randomness of isotope k ,⁵ stretches across the entire heterostructure. By splitting the integration domain into (i) the domain consisting of the 2D host material and (ii) the domain encompassing the substrate material, i.e.

$$\sum_{k \in \{\text{host}\} \cap \{\text{sub.}\}} -n_k \int F_k(r) d\mathbf{r} = \sum_{k \in \{\text{host}\} \cap \{\text{sub.}\}} -n_k \left[\int_{\text{host}} F_k(r) d\mathbf{r} + \int_{\text{sub.}} F_k(r) d\mathbf{r} \right], \quad (\text{S26})$$

we can express the coherence function as

$$\mathcal{L}(t) = \exp \left(\left[\sum_{i \in \{\text{host}\}} -n_i \int_{\text{host}} F_i(r) d\mathbf{r} \right] + \left[\sum_{j \in \{\text{sub.}\}} -n_j \int_{\text{sub.}} F_j(r) d\mathbf{r} \right] \right). \quad (\text{S27})$$

The goal is to express the coherence time of a heterostructure ($T_{2,\text{HS}}$) as a function of individual T_2 values of the 2D host ($T_{2,\text{host}}$) and substrate ($T_{2,\text{sub.}}$). The contribution of the 2D host is straightforward, as it follows

$$\begin{aligned} \sum_{i \in \{\text{host}\}} -n_i \int_{\text{host}} F_i(r) d\mathbf{r} &= \sum_{i \in \{\text{host}\}} - \left(\frac{t}{T_{2,\text{host},i}} \right)^{\eta_{\text{host}}} \\ &= - \left(\frac{t}{T_{2,\text{host}}} \right)^{\eta_{\text{host}}}. \end{aligned} \quad (\text{S28})$$

However, the substrate contribution is more nuanced. While $T_{2,\text{sub.}}$ can be predicted using the model of Kanai et al. (Eq. S17), the model assumes that the qubit is implanted deep in the substrate so that it interacts with the nuclear spin environment in all radial directions. Here, we consider a qubit that feels the effects of the substrate spin environment in the $-z$ direction only. Mathematically, this can be modelled by modifying the integration interval of the polar angle ϕ from $[0, \pi]$ to $[\pi/2, \pi]$ in spherical coordinates. In

particular,

$$\begin{aligned}
\sum_{j \in \{\text{sub.}\}} -n_j \int_{\text{sub.}} F_j(r) d\mathbf{r} &= \sum_{j \in \{\text{sub.}\}} -n_j \int_{\pi/2}^{\pi} \int_0^{2\pi} \int_0^{\infty} F_j(r) r^2 \sin(\theta) dr d\theta d\phi \\
&= \frac{1}{2} \sum_{j \in \{\text{sub.}\}} -n_j \int_0^{\pi} \int_0^{2\pi} \int_0^{\infty} F_j(r) r^2 \sin(\theta) dr d\theta d\phi \\
&= \frac{1}{2} \sum_{j \in \{\text{sub.}\}} -\left(\frac{t}{T_{2,\text{sub.},j}}\right)^{\eta_{\text{sub.}}} \\
&= -\frac{1}{2} \left(\frac{t}{T_{2,\text{sub.}}}\right)^{\eta_{\text{sub.}}}.
\end{aligned} \tag{S29}$$

By plugging Eqs. (S28) and (S29) into Eq. (S27), we obtain

$$\mathcal{L}(t) = \exp\left(-\left(\frac{t}{T_{2,\text{host}}}\right)^{\eta_{\text{host}}} - \frac{1}{2} \left(\frac{t}{T_{2,\text{sub.}}}\right)^{\eta_{\text{sub.}}}\right). \tag{S30}$$

Since we define the overall coherence function of the heterostructure in terms of $T_{2,\text{HS}}$ as

$$\mathcal{L}(t) \equiv \exp\left(-\left(\frac{t}{T_{2,\text{HS}}}\right)^{\eta_{\text{HS}}}\right), \tag{S31}$$

we obtain

$$\left(\frac{t}{T_{2,\text{HS}}}\right)^{\eta_{\text{HS}}} = \left(\frac{t}{T_{2,\text{host}}}\right)^{\eta_{\text{host}}} + \frac{1}{2} \left(\frac{t}{T_{2,\text{sub.}}}\right)^{\eta_{\text{sub.}}}. \tag{S32}$$

Here, $T_{2,\text{host}}$ for the 2D host material is given by Eq. (S21), and $T_{2,\text{sub.}}$ for the substrate can be determined using Kanai's model in Eq. (S17). The stretch exponent for the 2D host, η_{host} , is set to 1.06 as determined in [Supplementary Note 6](#) from fitting (see Eq. (S21)), and the stretch exponent for the substrate, $\eta_{\text{sub.}}$, is set to 1.5 since the material is 3D and we assume hyperfine interactions (see Eq. (S13)). We make the approximation

$$\left(\frac{1(s)}{T_{2,\text{HS}}(s)}\right)^{\eta_{\text{HS}}} \approx \left(\frac{1(s)}{T_{2,\text{host}}(s)}\right)^{\eta_{\text{host}}} + \frac{1}{2} \left(\frac{1(s)}{T_{2,\text{sub.}}(s)}\right)^{\eta_{\text{sub.}}} \tag{S33}$$

which leaves η_{HS} as the only fitting parameter. Note that the "(s)" in each term represents the time unit, ensuring that the units are matched between each term. By minimizing the relative absolute error, i.e.

$$\text{Error} = \sum_x \frac{|T_{2,\text{HS},x} - T_{2,\text{CCE},x}|}{T_{2,\text{CCE},x}} \tag{S34}$$

where the summation is performed over each heterostructure x , we obtain a fitted value of $\eta_{\text{HS}} = 1.35$.

Considerations

It is important to note that our model neglects decoherence effects from higher orders of nuclear spins (such as pairs), which can activate additional decoherence channels through e.g. flip-flop transitions. One can perhaps argue that flip-flop transitions in homonuclear spin baths are modeled “effectively” with the assumption of Gauss-Markov noise, where flip-flop transitions are a contributing factor. However, in Eq. (S26) within our model treatment of heterostructures, we completely neglect pair flip-flop transitions when the nuclear spin pair is composed of one nuclear spin in the 2D host material and another in the substrate. Such an interaction should manifest as an additional summand in Eq. (S26).

We assess the contributions from such pairs by dividing out the CCE-2 coherence functions of the 2D host material and substrate, which are calculated separately, from the full coherence function; in particular, we evaluate

$$\mathcal{L}_{\text{2D-substrate pairs}} = \frac{\mathcal{L}_{\text{Full}}}{\mathcal{L}_{\text{2D}}\mathcal{L}_{\text{substrate}}} \quad (\text{S35})$$

where $\mathcal{L}_{\text{Full}}$ is the CCE-2 coherence function from the full nuclear spin bath (including both the 2D host and substrate spin baths), and $\mathcal{L}_{\text{2D}}$ and $\mathcal{L}_{\text{substrate}}$ are the CCE-2 coherence functions from nuclear spin baths of the 2D host and substrate, respectively. Hence, $\mathcal{L}_{\text{Full}}$ includes the contributions of flip-flop transitions to decoherence from pairs consisting of one nuclear spin in the 2D host and another in the substrate, which are not included in $\mathcal{L}_{\text{2D}}$ nor $\mathcal{L}_{\text{substrate}}$. Since heteronuclear spin baths can be decoupled, it is important to consider heterostructural systems where a homonuclear spin bath spans both the 2D host material and substrate. Accordingly, we consider the following heterostructures: $\text{PdSO}_4/\text{SiO}_2$, $\text{NiSi}_2\text{Se}_4/\text{SiO}_2$, bilayer Si/SiO_2 , and monolayer Au/Au . In each case, the qubit is assumed to be in the 2D host layer. The first two systems are included in our dataset. Note that in each system considered, there is a homonuclear spin bath that spans both the 2D host material and substrate (^{17}O in $\text{PdSO}_4/\text{SiO}_2$, ^{29}Si in $\text{NiSi}_2\text{Se}_4/\text{SiO}_2$ and bilayer Si/SiO_2 , and ^{197}Au in monolayer Au/Au).

Our calculations show that $\mathcal{L}_{\text{Full}}$ is well-represented by $\mathcal{L}_{\text{2D}}\mathcal{L}_{\text{substrate}}$ (i.e. the product of the coherence functions from individual nuclear spin baths), especially in the early time regime (Figure S7). Our results therefore suggest that pairwise flip-flop transitions impose minimal decoherence channels for the spin qubit, validating our treatment of the host and substrate nuclear spin baths as two independently separable decoherence channels in Eq. (S26). $\mathcal{L}_{\text{2D-substrate pairs}}$ departs from unity in the later time regime, which is most severe in the case of monolayer Au/Au (Figure S7d) where the nuclear spin bath is densest (^{197}Au with 100% natural abundance). The trend makes sense intuitively, because a denser nuclear spin bath implies

more pairwise flip-flop transitions that can contribute to decoherence.

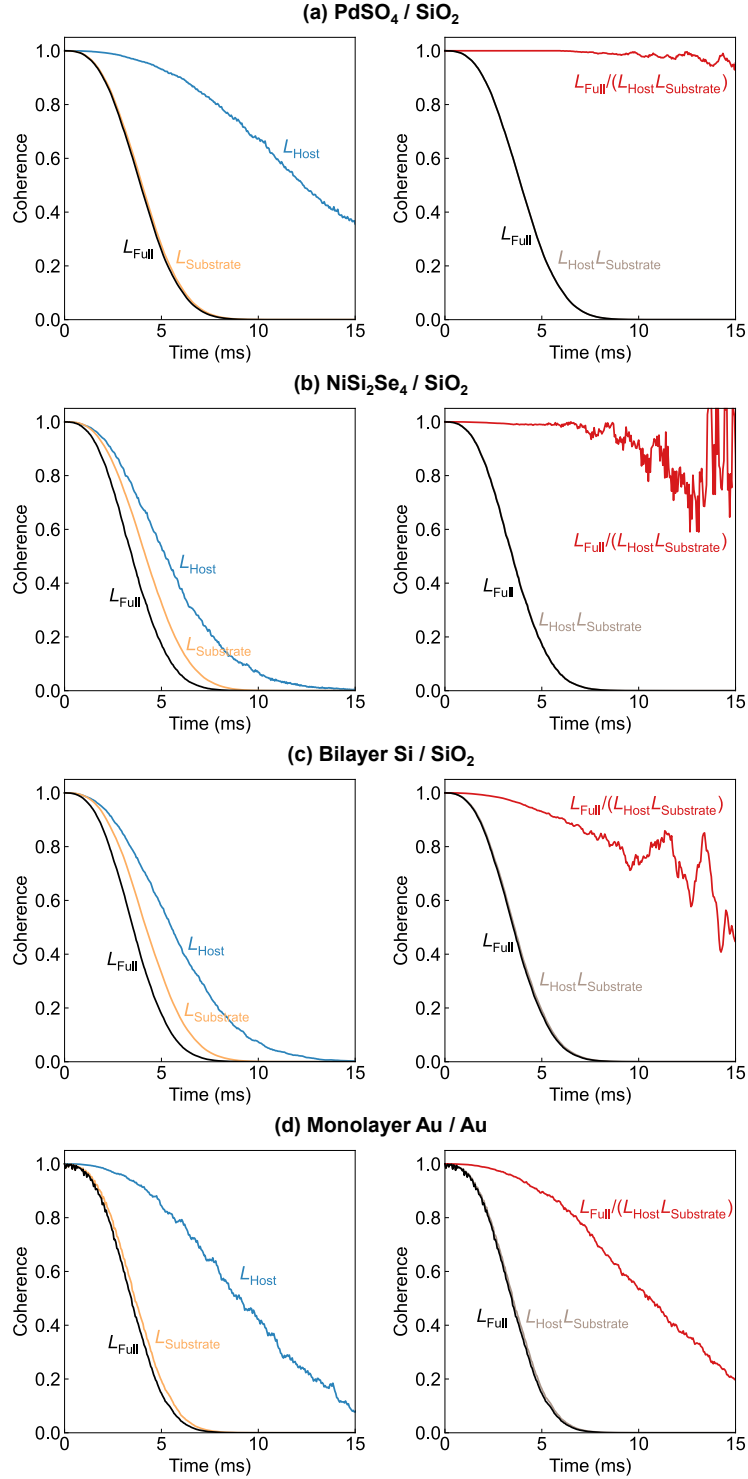


Figure S7: Coherence function of the full nuclear spin bath ($\mathcal{L}_{\text{Full}}$, black) compared to the coherence function from the host spin bath ($\mathcal{L}_{\text{Host}}$, blue) and substrate spin bath ($\mathcal{L}_{\text{Substrate}}$, orange). In most cases, $\mathcal{L}_{\text{Full}}$ can be approximated by the product of the individual coherence functions ($\mathcal{L}_{\text{Host}} \mathcal{L}_{\text{Substrate}}$, brown). The calculations are performed at a magnetic field of 5 T.

Supplementary Note 8: d -orbital splitting in a square-planar field

While specific spin defects are not identified in host materials in this study, one can envision the electronic structure that can be designed with an appropriate dopant. For example, there is a square-planar structural motif embedded in $\text{Au}_2\text{Se}_3\text{O}_{10}$ (AuO_4), $\text{Au}_2\text{Se}_2\text{O}_7$ (AuO_4), and PdSO_4 (PdO_4), which have the second, third, and fourth highest T_2 in our screening, respectively. Below, we show the electronic structure from d -orbital splitting in a square-planar configuration. By doping with an appropriate transition metal ion, it may be possible to realize a spin-1 qubit by occupying the e_g states with two unpaired electrons.

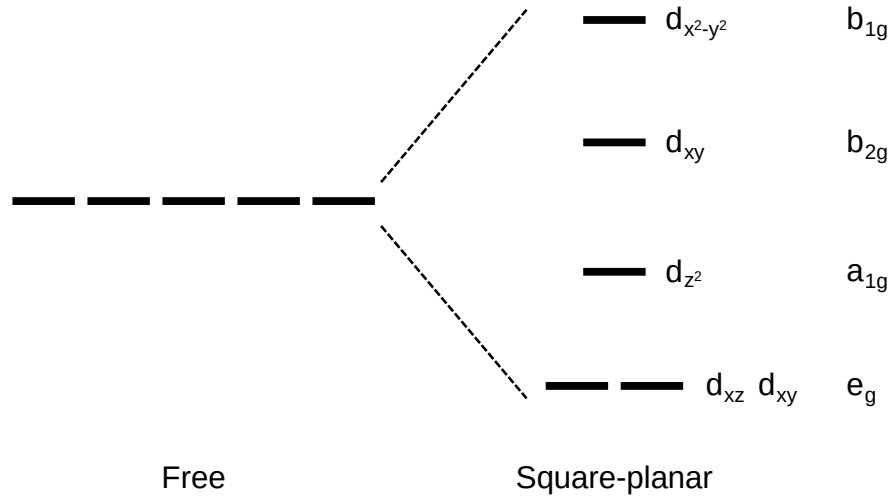


Figure S8: d -orbital splitting in a square-planar field. The square-planar structural motifs embedded in some high- T_2 materials predicted in the study, such as $\text{Au}_2\text{Se}_3\text{O}_{10}$, may enable the design of a spin-1 defect center by populating the e_g state with two unpaired electrons.

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