Hyperfine interaction for holes in quantum dots: $k \cdot p$ model

Paweł Machnikowski,$^1$ Krzysztof Gawarecki,$^1$ and Łukasz Cywiński$^2$

$^1$Department of Theoretical of Physics, Wrocław University of Science and Technology, 50-370 Wrocław, Poland
$^2$Institute of Physics, Polish Academy of Sciences, Warsaw, Poland

We formulate the multi-band $k \cdot p$ theory of hyperfine interactions for semiconductor nanostructures in the envelope function approximation. We apply this theoretical description to the fluctuations of the longitudinal and transverse Overhauser field experienced by a hole for a range of InGaAs quantum dots of various compositions and geometries. We find, in agreement with previous estimates, that the transverse Overhauser field is of the same order of magnitude as the longitudinal one and is dominated by the $d$-shell admixture to atomic states with only a minor correction from band mixing in all the cases. In consequence, the $k \cdot p$ results are well reproduced by a simple box model with the effective number of ions determined by the wave function participation number, as long as the hole is confined in the compositionally uniform volume of the dot, which holds in a wide range of parameters, excluding very flat dots.

I. INTRODUCTION

Hyperfine coupling between the spin of a hole localized in a self-assembled quantum dot (QD) and the nuclear spins of the atoms of the host materials has been a subject of intense experimental$^{1–8}$ and theoretical$^{6,9,10}$ investigations in recent years. The original reason for resurgence of interest in this topic was the prospect of using hole spins as qubits with long coherence times$^{1,2,8,11–14}$. This was motivated by the fact that dephasing of electron spins in QDs, being an obstacle to their applications as qubits with long coherence times$^{1,2,8,11–14}$, is dominated by their hyperfine (hf) interaction with the nuclear spins of the host material$^{15–19}$, and the hole-nucleus coupling was expected to be much weaker than the electron-nucleus one$^{9,10}$. The dominating mechanism in the case of electron hf coupling is the contact interaction$^{15}$. On the contrary, the holes have very small contribution of $s$ states in their wave functions, and therefore interact with nuclei by weaker (approximately by one order of magnitude$^9$) and much more subtle dipole couplings, which are sensitive to the details of their atomic (or Bloch) wave functions. Due to non-$s$ character of hole Bloch states, a strong anisotropy of this interaction with respect to growth axis was also predicted$^9,10$. This general expectation was confirmed by experiments, showing that the Overhauser field exerted by the nuclei on the hole is about an order of magnitude smaller than the one experienced by the electron in the same dot$^{1–5}$. These results opened the way for using hole spin qubits in applications, such as creation of long-distance entanglement of hole spins$^{13}$, in which their enhanced coherence time and good coupling to photons (holding for both holes and electrons in self-assembled QDs$^{20,21}$) was helpful.

However, despite the qualitative agreement between early predictions and experimental results, a number of controversies and open questions arose. While the longitudinal (along the QD growth axis) hole-nuclear interaction qualitatively agreed with early predictions, large discrepancies in strength of transverse hyperfine interactions were reported, with estimated values of transverse coupling ranging from the same order of magnitude$^3$ to less than $1\%$ of the longitudinal one$^8$. The origin of the transverse interactions is also controversial: while initially heavy-light hole mixing was invoked$^{3,9,10}$ to explain its finite value, the presence of finite admixture of $d$-symmetry states in states forming the top of valence band in relevant III-V materials was later suggested to play a significant, or even possibly dominant, role$^6$. Such a substantial admixture of atomic $d$ states in the valence band Bloch functions is in accordance with earlier theoretical results$^{22–25}$.

It is important to note that with transverse hole hyperfine coupling being much smaller than the longitudinal one (which, in turn, is lower by an order of magnitude than the coupling for an electron), the coherence time of hole spin polarized along the growth axis can be significantly enhanced by application of large transverse magnetic field$^8$. Understanding of physical origin of the transverse coupling is thus important, as it would possibly allow for design of QDs (by varying composition/shape/strain etc.) with the best possible hole spin coherence properties.

It should also be noted that interpretation of most experiments related to physics of carrier and nuclear spins in QDs relies on simplified models of carrier envelope wave functions (e.g. assuming the same envelope shapes for holes and electrons) and all the multi-band effects (including the degree of heavy-light hole mixing), and their relation to QD shape. This includes works on carrier spin coherence, that apart from nuclear effects show influence of charge noise coupling to spin via electric-field dependent $g$-factors$^{11,14}$, creation of dynamic nuclear polarization$^{4,5}$, and optical detection (through changes in Overhauser field-induced spin splitting of electron and holes) of nuclear magnetic resonance of different species of nuclei present in the dot$^{26}$. While such experiments were used to obtain new information on structural properties and strain distribution in QDs$^{26}$, the simplicity of some of the above-mentioned assumptions casts a certain degree of doubt on the interpretation of measurement results.

The current state of the art in the theoretical model-
ing of self-assembled semiconductor structures is to use either atomistic methods \cite{27-29} or multi-band \textbf{k}·\textbf{p} theories in the envelope function approximation \cite{30,31}. The latter has found a vast range of applications due to its relatively low computational cost and high versatility. It offers reliable information on the wave function geometry and band mixing and allows one to quantitatively relate the observed spectral features to fine details of the nano-system morphology and composition. It can be used not only to compute the carrier states and the resulting optical transitions \cite{32}, but also to model carrier-phonon couplings \cite{33} and to evaluate the spin-related properties, including \textit{g}-factors \cite{34,35}, the effects of spin-orbit coupling \cite{31,36}, as well as phonon-induced spin relaxation and dephasing \cite{37,38}. Therefore, in terms of quantitative accuracy, a simple approach to hyperfine interactions lags behind the current standards in the modeling of carrier wave functions in semiconductor nanostructures and is not on a par with the sophistication of experimental techniques used for the measurements of the relevant quantities. It therefore seems useful to develop a theory that would allow one to combine the hyperfine interaction with realistic modeling of wave functions. Such a more general and accurate theory may be useful in systems with compositional inhomogeneity and controllable carrier localization, like double QDs, or with strong in-plane anisotropy, where band mixing is relatively stronger \cite{39}.

The goal of this paper is to revisit the problem of calculation of the anisotropic Overhauser field acting on a hole spin while employing a detailed realistic description of carrier states in QDs. We derive a theoretical description of hyperfine coupling for a carrier confined in a self-assembled semiconductor QD based on the multi-band wave function obtained from the \textbf{k}·\textbf{p} theory in the envelope function approximation, taking into account d-wave admixture in the valence-band states. In this way, we provide a model of the hyperfine interaction compatible with the standard \textbf{k}·\textbf{p} modeling of carrier states, which opens the way towards combining the effects of hyperfine coupling with reliable modeling of other characteristics of the QD system. As an application of the formalism, we calculate the rms fluctuations of the longitudinal and transverse Overhauser field in InGaAs/GaAs QDs and compare the contributions to the transverse field fluctuations from band mixing and d-wave admixture to valence band states.

The paper is organized as follows. In Sec. II we derive the general 8-band \textbf{k}·\textbf{p} Hamiltonian for hyperfine interactions. Next, in Sec. III we apply this formalism to the fluctuations of the Overhauser field felt by a hole in a QD. Sec. IV summarizes our findings. Technical derivations are collected in the Appendix.

II. THE MULTI-BAND HYPERFINE HAMILTONIAN

The hyperfine Hamiltonian describes the interaction of the carrier with all the nuclei (labeled by $\alpha$ and located at $\mathbf{R}_\alpha$),

\[ H = 3E_{hf} \sum_\alpha \zeta_\alpha \mathbf{A}(r - \mathbf{R}_\alpha) \cdot \mathbf{I}_\alpha / \hbar, \]

where

\[ E_{hf} = \frac{2\mu_0}{3\pi} \mu_B \mu_N a_B^{-3} = 0.5253 \mu eV, \]

$\mu_B$ and $\mu_N$ are Bohr and nuclear magnetons, respectively, $a_B$ is the Bohr radius, $\mu_0$ is the vacuum permeability, $I_\alpha$ is the nuclear spin, $\zeta_\alpha$ defines the nuclear magnetic moment for a given nucleus via $\mu_\alpha = \zeta_\alpha \mu_N I_\alpha$, and

\[ \mathbf{A}(r) = \frac{a_B^3}{4\hbar} \left[ \frac{8\pi}{3} \delta(r) S + \frac{L}{r^3} + \frac{3(\mathbf{r} \cdot \mathbf{S})\mathbf{r} - \mathbf{S}}{r^3} \right]. \]

Within the envelope function approach to the \textbf{k}·\textbf{p} theory, the wave functions are decomposed into contributions from various bands $\lambda$ with $\Gamma$-point Bloch functions $u_\lambda(r, s)$,

\[ \Psi_\mu(r, s) = \sum_\lambda \psi_{\mu,\lambda}(r) u_\lambda(r, s), \]

where the envelopes $\psi_{\mu,\lambda}(r)$ are assumed to vary slowly in space (as compared to the lattice constant) and $s$ denotes the spin projection. Most commonly, an 8-band model is used \cite{36}, explicitly representing two subbands of the conduction band (belonging to the $\Gamma_{6c}$ representation of the bulk crystal) and six subbands in the valence band (four-dimensional $\Gamma_{8c}$ and two-dimensional $\Gamma_{7c}$), with the coupling to other bands represented by effective terms resulting from perturbation theory. The eight envelope wave functions $\{\psi_{\mu,\lambda}(r)\}$ are commonly thought of as an 8-component “pseudo-spinor”. Consequently, the Hamiltonian (or any other operator) in the envelope-function \textbf{k}·\textbf{p} theory can be considered an 8 × 8 array of operators $H_{\lambda',\lambda}$ in the coordinate representation, such that any matrix element of the original Hamiltonian is given by

\[ \langle \nu | \mathbf{H} | \mu \rangle = \sum_{\lambda',\lambda} \int d^3r \psi^*_{\nu,\lambda'}(r) H_{\lambda',\lambda} \psi_{\mu,\lambda}(r). \]

The goal of this section is to apply the envelope function approximation [Eq. (3)] to the hyperfine Hamiltonian (1) and to write it in the form consistent with Eq. (4).

Starting from Eq. (2) and using Eq. (3), the matrix elements of $A_i$ are

\[ \langle \nu | A_i(r - R_\alpha) | \mu \rangle = \sum_{\lambda',\lambda} \sum_{ss'} \int d^3r \psi^*_{\nu,\lambda'}(r) u_{\lambda'}^*(r, s') A_i, s' s (r - R_\alpha) \psi_{\mu,\lambda}(r) u_{\lambda}(r, s), \]

\[ \times A_i, s' s (r - R_\alpha) \psi_{\mu,\lambda}(r) u_{\lambda}(r, s), \]
where $A_{\alpha}(r)$ denotes the matrix elements of $A_i(r)$ with respect to spin states. The Bloch functions are decomposed into parts localized around the anion (A) and cation (C), that are assumed normalized and non-overlapping,

$$u_{\alpha}(r,s) = \sum_{i=A,C} a_{\alpha}^{(i)}(r,s),$$

where $a_{\alpha}^{(i)}$ are the contributions of the anionic and cationic atomic orbitals to a given band. Next, we split the space into primitive cells, which are further divided into two parts surrounding the anion and the cation. The integration over the whole space is then performed as integration over the surrounding of each ion and summation over all the ions. We use the fact that the envelope varies slowly, so that in the vicinity of each ion it can be approximated by its value at the ion position $R$. In this way we transfer Eq. (5) into

$$\langle \nu | A_i(r-R_\alpha) | \mu \rangle = v \sum_{\alpha',\ldots} \psi_{\nu,\ldots}(R_{\alpha'}) A_{\alpha,\ldots}^{\nu,\ldots}(R_{\alpha'}) u_{\alpha}(r,s),$$

with

$$A_{\alpha,\ldots}^{\nu,\ldots} = \frac{1}{v} \sum_{\alpha' s'} \int d^3r \, u_{\alpha'}^{s'}(r,s') A_{\alpha,s'}(r-R_{\alpha}) u_{\alpha}(r,s),$$

where $v$ is the volume of the primitive crystal cell and $V_\alpha$ denotes the volume surrounding the ion $\alpha$ (the arbitrariness in choosing this volume is unimportant in view of the strong localization of Bloch functions around the ions). Since the variation of the envelope functions is slow, the summation in Eq. (6) realizes a coarse-grained integration over the whole space. Thus, Eq. (6) brings matrix elements of the hyperfine Hamiltonian, Eq. (1), to the form of Eq. (4) with

$$H_{\lambda,\ldots}(r) = 3E_{hf}^{\lambda,\ldots} \sum_{\alpha',\ldots} \delta(r-R_{\alpha}) \zeta_{\alpha} A_{\alpha,\ldots}^{\nu,\ldots} \cdot \mathbf{I}_\alpha / \hbar,$$

In order to evaluate Eq. (7) one needs a model of the Bloch functions. Following Ref. 9, we choose to represent them as combinations of normalized hydrogen-like functions $f_{lm}^{(i)}(r)$ with definite rotation symmetry ($l = s,p,d$), characterized by the orbital exponents $\xi_i^{(a)}$ that depends on the nuclear species occupying the site $\alpha$. Thus,

$$u_{\alpha}^{(i)}(r,s) = \sqrt{v} \sum_{lm} c_{im}^{(i)} f_{lm}^{(i)}(r-r_1),$$

where $l = 0,1,2$, $m = -l,\ldots,l$. The valence band Bloch functions are composed of $p$ and $d$ atomic orbitals, weighted by the amplitudes $\alpha_p$ and $\alpha_d$, respectively, with $|\alpha_p|^2 + |\alpha_d|^2 = 1$. Here we suppress the principal quantum number $n$ since, in a typical III-V compound, it happens to be uniquely determined for a given element by $l$ (e.g., only $3d$, $4s$, and $4p$ states are involved for Ga and As, while $4d$, $5s$, and $5p$ are relevant for In). The coefficients $c_{im}^{(i)}$ for purely $p$-band ($l = 1$) states can be found from angular momentum addition and are widely available in the literature related to the $k \cdot p$ method. The extension to the $d$ admixture follows immediately from the explicit form of the basis functions of the $F_2$ representation of the $T_d$ point group, as given in Ref. 6.

The matrix element in Eq. (8) has two contributions: the local, or short-range (SR) one, from the surrounding of the ion in question ($\alpha' = \alpha$) and the long-range (LR) one, from all the other ions in the crystal (including the neighboring cations for an anion and vice-versa). The LR contribution has been estimated to be negligible.

In the following we only take into account the SR contribution.

The detailed derivation of the SR contributions, which systematically extends the existing theoretical description to multi-band wave functions, is given in the Appendix. The resulting matrix elements $A_{\alpha,\ldots}^{\nu,\ldots}$ must have appropriate transformation properties, hence they can be expressed by the standard matrices used to define point group invariants when constructing the $k \cdot p$ theory. In order to use this convenient notation we split the array $\{H_{\lambda,\ldots}\}$ into blocks corresponding to the three irreducible representations spanning the 8-band $k \cdot p$ model,

$$H = \begin{pmatrix} H_{66} & H_{68} & H_{67} & H_{77} \\ H_{68} & H_{88} & H_{87} & 0 \\ H_{77} & H_{78} & H_{77} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix},$$

with

$$H_{bb'} = H_{b'b} = E_{hf} v \sum_{\alpha} \delta(r-R_{\alpha}) a_{\alpha}^{(x)\ast} a_{\alpha}^{(x)} \zeta_{\alpha} \xi_{8,\alpha} T_{bb'}^{(x)}$$

(the index $b$ labels blocks), and find

$$\tilde{H}_{66}^{(x)} = \sigma \cdot \mathbf{I}_\alpha / \hbar,$$

$$\tilde{H}_{68}^{(x)} = \left( -\frac{8}{5} \tilde{M}_p^{(x)} + \frac{39}{7} \tilde{M}_d^{(x)} \right) \mathbf{J} \cdot \mathbf{I}_\alpha / \hbar,$$

$$\tilde{H}_{77}^{(x)} = \left( -4 \tilde{M}_p^{(x)} + \frac{5}{7} \tilde{M}_d^{(x)} \right) \mathbf{J} \cdot \mathbf{I}_\alpha / \hbar,$$

$$\tilde{H}_{88}^{(x)} = \left( \frac{9}{\sqrt{5}} \tilde{M}_d^{(x)} + T_{xy} \mathbf{I}_{x,x} + T_{yz} \mathbf{I}_{y,y} + T_{zx} \mathbf{I}_{z,z} \right) / \hbar,$$

$$\tilde{H}_{66}^{(x)} = 0,$$

$$\tilde{H}_{77}^{(x)} = -\sqrt{3} \left( \tilde{M}_p^{(x)} - \frac{15}{7} \tilde{M}_d^{(x)} \right) \mathbf{T} \cdot \mathbf{I}_\alpha / \hbar,$$

where $\tilde{M}_p^{(x)} = |\alpha_p|^{2} \tilde{M}_p^{(x)}$, $\tilde{M}_d^{(x)} = |\alpha_d|^{2} \tilde{M}_d^{(x)}$, the dimensionless quantities $\tilde{M}_p^{(x)}$, $\tilde{M}_d^{(x)}$ characterize the geometry of the atomic functions and are explicitly defined.
in the Appendix, \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) are Pauli matrices, \( \mathbf{J} = (J_x, J_y, J_z) \) are the matrices of the 4-dimensional \( (j = 3/2) \) irreducible representation of angular momentum, \( \mathbf{J} = (J_x^3, J_y^3, J_z^3) \),

\[
T_x = \frac{1}{3\sqrt{2}} \begin{pmatrix} -\frac{\sqrt{3}}{2} & 0 & 1 & 0 \\ 0 & 1 & 0 & \sqrt{3} \end{pmatrix}, \\
T_y = -\frac{i}{3\sqrt{2}} \begin{pmatrix} \frac{\sqrt{3}}{2} & 0 & 1 & 0 \\ 0 & 1 & 0 & -\sqrt{3} \end{pmatrix}, \\
T_z = \frac{\sqrt{2}}{3} \begin{pmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{pmatrix},
\]

and \( T_{ij} = T_i J_j + T_j J_i \). Here the equation for \( \tilde{H}_{Sv8v} \) reproduces the result of Ref. 6.

From Eq. (12b) it is clear that for a purely hh state the only flip-flop terms appear as a result of \( d \)-shell admixture via the \( J_i^3 \) terms that reflect the lowered symmetry of the crystal as compared to the full rotation group. As we show in the Appendix, these terms originate from the spin part of the dipole hyperfine coupling (the last term in Eq. (2)). Inter-band terms in the Hamiltonian lead also to flip-flop processes induced by band mixing but, as we will see below, this effect is much weaker.

## III. HYPERFINE COUPLING FOR THE HEAVY-HOLE GROUND STATE

In this section we apply the general formalism of Sec. II to the ground state Zeeman doublet of the nominally heavy hole state in a range of self-assembled QDs with varying size, shape and composition. We characterize the fluctuations of the Overhauser field felt by the hole that is the key factor determining the hyperfine-induced spin dephasing.

### A. QD model and wave functions

The envelope functions for the QD ground state are computed for a few series of QD structures with the 8-band \( k \cdot p \) theory. In all the cases the composition of the QD is uniform and corresponds to the stoichiometric formula \( \text{In}_x\text{Ga}_{1-x}\text{As} \). The QD is placed on a wetting layer of the same composition and thickness equal to the GaAs lattice constant \( a = 0.565 \) nm.

We account for the strain within continuous-elasticity approach\(^4^1\). We take into account the piezoelectric potential, up to the second order in polarization\(^4^5\). The magnetic field enters via Peierls substitution within the gauge invariant scheme, described in detail in Ref. 46. The detailed description of the model as well as parameters used in computations are given in Ref. 31.

Table I lists the values of the parameters relevant for the modeling of Bloch functions and hyperfine couplings (see Sec. II and Appendix): nuclear spin quantum numbers, \( \zeta \) coefficients and relative abundances \( r \) for the nuclei of interest, the atomic wave function exponents \( \xi \) and the resulting \( M \) parameters, as well as the \( d \)-state admixture amplitude assumed in the calculations. We assume equal distribution of the wave function between the anion and the cation in all the bands, that is, \( a_\alpha(\lambda) = 1/\sqrt{2} \) for each ion \( \alpha \) and for each band \( \lambda \).

| \( ^{69}\text{Ga} \) | \( ^{71}\text{Ga} \) | \( ^{113}\text{In} \) | \( ^{115}\text{In} \) | \( ^{75}\text{As} \) |
|---|---|---|---|---|
| \( T \) | 3/2 | 3/2 | 9/2 | 9/2 | 3/2 |
| \( \zeta \) | 1.344 | 1.708 | 1.227 | 1.230 | 0.959 |
| \( r \) | 0.604 | 0.396 | 0.0428 | 0.9572 | 1 |
| \( \xi_s \) | 1.7667 | 1.9023 | 2.2360 |
| \( \xi_p \) | 1.5540 | 1.6940 | 1.8623 |
| \( \xi_d \) | 5.0311 | 4.2354 | 5.7928 |
| \( M_p \) | 0.05687 | 0.05885 | 0.04815 |
| \( M_d \) | 0.3849 | 0.1839 | 0.2898 |
| \( M_{sd} \) | 0.06145 | 0.03014 | 0.03979 |
| \( \alpha_{sd} \) | 0.41 | 0.69 | 0.17 |

### B. Effective Hamiltonian

We find the effective Hamiltonian describing the hyperfine interactions in the heavy hole ground state by projecting Eq. (1) onto the two-dimensional space of the ground-state doublet. We denote the eigenstates in the ground state doublet (as resulting from the \( k \cdot p \) diagonalization) by \( \ket{\uparrow} \) and \( \ket{\downarrow} \) (hence the two basis states are defined with respect to the spin quantization axis) and define operators \( \mathbf{\Sigma} \) corresponding to Pauli matrices in this two-dimensional subspace, \( \mathbf{\Sigma} = \ket{\uparrow}\!\!\!\langle\uparrow| - \ket{\downarrow}\!\!\!\langle\downarrow| \) etc. The Hamiltonian given in Eq. (1) is linear in the nuclear spins, hence its projection on the two-dimensional subspace can be written as

\[
H = \frac{1}{2} \sum_{\alpha} \sum_{ij} \mathcal{H}^{(\alpha)}_{ij} (I_{\alpha,i}/\hbar) \mathbf{\Sigma}_j , 
\] (13)

where

\[
\mathcal{H}^{(\alpha)}_{ij} = 3E_{\text{hf}}\zeta_\alpha \xi_{s,\alpha}^3 \text{Tr} \left[ A_i (r - R_\alpha) \mathbf{\Sigma}_j \right] .
\]

This Hamiltonian has the form of a Zeeman Hamiltonian,

\[
H = \frac{1}{2} \mathbf{\hbar} \cdot \mathbf{\Sigma} ,
\]

with the quantity \( \hbar \), defining the Overhauser field, with components given by

\[
h_j = \sum_{\alpha} \sum_i \mathcal{H}^{(\alpha)}_{ij} I_{\alpha,i}/\hbar .
\]

We assume here that the nuclei are in thermal state without any dynamical polarization. Except for unrealistically low temperatures, this means that the nuclear density matrix is maximally mixed. The mean square of a
given component of $\mathbf{h}$ is then given by

$$
\langle h_{ij}^2 \rangle = \sum_{\alpha, i} \sum_{\alpha', i'} \mathcal{H}^{(\alpha)}_{ij} \mathcal{H}^{(\alpha')}_{ij} \langle I_{\alpha, i} I_{\alpha', i'} \rangle / \hbar^2
$$

$$
= \frac{1}{3} \sum_{\alpha} I_{\alpha} (I_{\alpha} + 1) \sum_{i} \left( \mathcal{H}^{(\alpha)}_{ii} \right)^2,
$$

where the last equality assumes that angular momenta of different nuclei as well as different components of nuclear spin are uncorrelated.

In the simplest approximation, one considers a purely heavy-hole wave function which occupies a region of uniform composition and is the same for both spin orientations. Then, by direct inspection of Eq. (12b) one finds

$$
\mathcal{H}^{(\alpha)}_{ii} = 2eE_{\text{hh}} |\psi(R_{\alpha})|^2 \zeta_{x_{s, \alpha}} c^3 \mathcal{M}^{(\alpha)}_{i},
$$

$$
\mathcal{H}^{(\alpha)}_{ij} = 0, \quad i \neq j,
$$

where

$$
\mathcal{M}^{(\alpha)}_{x} = \mathcal{M}^{(\alpha)}_{y} = -\frac{9}{7} M_{d}^{(\alpha)},
$$

$$
\mathcal{M}^{(\alpha)}_{z} = -\frac{12}{5} M_{p}^{(\alpha)} + \frac{18}{7} M_{d}^{(\alpha)}.
$$

Since $M_{d}^{(\alpha)}$ depends only on the species of the ion $\alpha$ and $\psi(R)$ changes slowly, one can write for the ternary compound In$_x$Ga$_{1-x}$As

$$
\langle h_{ij}^2 \rangle = 4E_{\text{hh}}^2 v \int d^3R |\psi(R)|^4 \sum_i \frac{I_i(I_i + 1)}{3} q_i \left( \zeta_{s, i} c^3 \mathcal{M}_{i}^{(\alpha)} \right)^2
$$

where $i$ runs through all the isotopes, $q_i = r_i x$ for In isotopes, $q_i = r_i (1 - x)$ for Ga isotopes and $q_i = 1$ for As. The quantity

$$
N = \left[ v \int d^3R |\psi(R)|^4 \right]^{-1}
$$

is the effective number of the primitive cells encompassed by the wave function (the inverse participation number) which links the presented theory to the box model in which the wave function is considered constant, with the value $1/\sqrt{vN}$ over a volume of $N$ unit cells.

C. Results and discussion

In this section we study the characteristic strength of the coupling to longitudinal and transverse fluctuations of the Overhauser field felt by a hole in the QD (nominally heavy-hole) ground state. According to the definitions in Sec. III B, the $z$ axis is the spin quantization axis, hence the notions of ‘longitudinal’ and ‘transverse’ are related in the usual way to the field orientation. All the results are averages of 10 repetitions in order to account for the random alloying and isotope distribution, resulting in a standard deviation of the numerical result on the order of 1% of the average value. Transverse fluctuations are calculated as the average of fluctuations in two perpendicular directions, $\langle h_{ij}^2 \rangle^{1/2} = (\langle h_{x}^2 + h_{y}^2 \rangle)/2^{1/2}$.

Fig. 1 shows the results for four series of structures with different size and composition. The magnetic field is oriented here in the growth direction (Faraday configuration), hence the $z$ axis is along the symmetry axis of the structure.

In Fig. 1(a) we study cylindrically symmetric lens-shaped QDs with base radius $2a$ and height $h = 7a = 3.96$ nm, and with uniform composition In$_x$Ga$_{1-x}$As, where the indium content $x$ changes from 0.1 to 1. Without $d$-state admixture to the valence band and without band-mixing, a heavy hole couples only to longitudinal hyperfine field. Band mixing induces weak coupling to transverse field (blue open circles), up to a few percent of the longitudinal one. A much stronger coupling, comparable to the longitudinal one, appears as a result of $d$-state admixture (full red circles). The strong dependence on the In content results from the combination of the large nuclear angular momentum of this element as compared to Ga and increasing localization in indium-rich QDs (the inverse participation number $N$ decreases from $208 \cdot 10^3$ to $52 \cdot 10^3$ as $x$ grows from 0.1 to 1). This dependence is much weaker in the case of transverse cou-
sampling induced purely by band mixing. The grey solid lines show the results obtained from Eq. (14). In order to relate our multi-band numerical wave functions to the simple theory we define here the inverse participation number as

\[ N' = \left[ V \int d^3 R \left| \sum_\lambda |\psi_\lambda (R)|^2 \right|^2 \right]^{-1} \]

and average the result over the two hh states. The agreement is very good, validating the box model with the inverse participation number as the effective number of primitive cells.

Fig. 1(b) presents results for a series of QDs with identical compositions \( x = 0.75 \), starting from the geometry as in the previous case and then uniformly scaling the size up by a factor up to 2. In Fig. 1(c) the lateral size of the QD is kept fixed as in (a) and the height \( h \) is varied. In Fig. 1(d) the QD is made elliptic by elongating the QD shape by a fixed factor along the \((100)\) crystallographic axis. In all these cases the general dependence on the geometry qualitatively follows the prediction of the box model with the effective field fluctuations decreasing with the growing system size. Quantitatively, however, the fluctuations of the Overhauser field only approximately follow the expected scaling as \( 1/\sqrt{V} \), which is due to the fact that the wave function shrinks slower than the QD when the size of the latter is reduced. In Fig. 1(c) one can see discrepancy between the numerical values and the predictions of the box model for very flat QDs. This results from the leakage of the wave function to the indium-free barrier.

For the amplitudes of the \( d \)-state admixtures used here (extracted from the data in Ref. 6), the magnitudes of the longitudinal fluctuations with and without the \( d \)-state admixture are very similar, which is, however, a coincidence. The dependence of the Overhauser field fluctuations on the assumed magnitude of \( d \)-shell admixture is shown in Fig. 2, where we present the results of calculations with the \( d \)-shell admixture magnitude for the nuclear species \( i \) set to \( |\alpha_d^{(i)}|^2 = y|\alpha_d^{(0)}|^2 \), where \( \alpha_d^{(0)} \) is the value extracted from the experiment and used in the calculations presented above, and \( 0 < y < 1 \). The dependence is non-monotonic. In particular, \( y \approx 0.5 \) corresponds to mutual compensation of the \( p \) and \( d \) contributions to the coupling to indium ions, which dominate the overall effect due to their large nuclear momentum. As a result, the longitudinal fluctuations of the effective field are suppressed.

In the strongly confined self-assembled structure the volume occupied by the wave function depends very weakly on the magnitude and orientation of the magnetic field. Therefore, one expects that the fluctuations of the Overhauser field will not depend on the magnetic field, except for the obvious redefinition of components, according to the field orientation. In particular, for the Voigt geometry with the field along the \((100)\) direction, the components of the Overhauser field in terms of the components for the Faraday configuration are

\[ h_x^{(V)} = -h_z, \quad h_y^{(V)} = h_y, \quad h_z^{(V)} = h_x. \]

The prediction for the fluctuation of these components obtained from the previous numerical results by such a simple rotation of components are shown with grey lines in Fig. 3(a). The red symbols represent the actual results obtained from numerical computation with the field of 8 T in the Voigt geometry, showing perfect agreement with the prediction. In addition, we performed computations in the Voigt geometry at \( B = 1 \) T, shown with crosses in Fig. 3. It is clear that the results do not depend on the field magnitude. In Fig. 3(b) we show analogous results from a model assuming no \( d \)-shell admixture. As expected, the transverse and fluctuations are now much stronger than the longitudinal ones, which...
immediately follows from the data in Fig. 1(a) (open symbols) by rotation of components, with the component in the growth direction much larger than the in-plane ones.

IV. CONCLUSIONS

We have derived the 8-band $k \cdot p$ Hamiltonian for hyperfine interactions. This offers a general formalism that allows one to include realistic multi-band carrier wave functions, as obtained from $k \cdot p$ computations, in the calculation of hyperfine couplings. Using this formalism, we have studied the effect of fluctuations of the nuclear spin polarization on a hole in the ground state of an InGaAs QD for a range of realistic shapes and sizes, taking into account an admixture of atomic $d$ orbitals to the valence band Bloch functions as well as band mixing. One of the main results is the observation that in a wide range of dots shapes and sizes, the realistic description of carrier states, taking into account band mixing, envelope functions leakage into the barrier, etc., has little influence on states, taking into account band mixing, envelope functions leakage into the barrier, etc., has little influence on transitions leakage into the barrier, etc., has little influence on until now in the latter case.

For the transverse fluctuations of the Overhauser field, we have confirmed the relatively small effect of band mixing as compared to the $d$-state admixture. The latter may lead to transverse fluctuations on the same order of magnitude as the longitudinal ones. The dependence of the longitudinal fluctuations on the amount of $d$ admixture is strong and non-monotonic. In the light of the fact that a large variability in the magnitude of both transverse and longitudinal fluctuations was reported in experiments, these results suggest the need for careful examination of dependence of the magnitude of the $d$-state admixtures to wave functions localized close to cation and anion cores, as a function of indium content (and possibly strain) in InGaAs/GaAs QDs.

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Appendix A: Derivation of the matrix elements of the hyperfine coupling

The main purpose of this Appendix is to rigorously derive the matrix elements of the short-range multi-band Hyperfine Hamiltonian as given in Eqs. (12a)–(12f). In the following, we will focus on one selected nucleus located at $\mathbf{R}_0$ and the index $\alpha$ will be suppressed. The three contributions to $\mathbf{A}$ in Eq. (2) will be denoted, respectively, as $\mathbf{A}_c$ (the contact interaction), $\mathbf{A}_o$ (the orbital part of the dipole interaction), and $\mathbf{A}_s$ (the spin part of the dipole interaction).

For the general calculations to be performed, it is convenient to use spherical tensor representation of various vectorial and tensorial quantities that appear in the derivations. Before we proceed do the technical derivations let us note that this is a natural language for discussing the hyperfine spin-flip selection rules. The essential part of the hyperfine Hamiltonian in Eq. (1) is

$$A(r - R_0) \cdot I = -\sqrt{3} \sum_{q_1, q_2} \langle 1; 1; q_1, q_2 | 1; 0, 0 \rangle A^{(1)}_{q_1}(r - R_0) A^{(1)}_{q_2},$$

where the upper index denotes the rank of the tensor and, at the same time, distinguishes the spherical tensor components from the cartesian ones, $\langle j_1, j_2; m_1, m_2 | j_1, j_2; j, m \rangle$ is the Clebsch-Gordan coefficient and the spherical components of any vector $\mathbf{V}$ are defined in the standard way,

$$V^{(1)}_{0,1} = V_z, \quad V^{(1)}_{\pm 1} = \mp V_z - i V_y \sqrt{2} = \mp \frac{1}{\sqrt{2}} V_{\pm}.$$

The range of $q_1, q_2$ is not given explicitly upon assumption that ill-defined Clebsch-Gordon coefficients are 0. Explicitly, $\mathbf{A} \cdot I = A^{(1)}_c J^{(1)}_c - A^{(1)}_o J^{(1)}_o - A^{(1)}_s J^{(1)}_s = A_z J_z + (A_+ J_+ + A_- J_-)/2$. The $q = 0$ term thus corresponds to the Ising coupling, which leads to Zeeman splitting of electron or hole states in the effective field generated by the nuclei (the Overhauser field). The $q = \pm 1$ terms account for spin flip-flop processes, in which the carrier exchanges its spin with the nucleus. In the simplest picture of hole states with definite angular momentum and composed exclusively of $p$ orbitals, the contact part $\mathbf{A}_c$ does not contribute to valence band hyperfine coupling due to vanishing $p$-type wave functions at the position of the nucleus. The other two terms can only contribute to diagonal terms, since the vector operator $\mathbf{A}$ cannot couple states with $m_j = \pm 3/2$, that is, differing by $|\Delta m_j| = 3$. Hence, in this single-band approximation, only the Ising term appears for heavy holes. However, symmetry reduction in a nanostructure modifies this simple picture by mixing the states belonging to different representations of angular momentum due to band mixing as well as by admixing $d$-shell atomic orbitals to valence band Bloch functions.

For the derivations we note that, by comparing Eq. (8) with Eq. (11), the Hamiltonian blocks $\tilde{H}_{p,b}$ contain
grouped elements $3A_{\lambda\lambda} \cdot I/(\hbar a^{(X)^*}a^\lambda)$. We will now derive these elements for each of the three contributions to the hyperfine Hamiltonian.

\[ \text{a. The contact part} \]

The contact part, i.e., the first term in Eq. (2), has contributions only from the conduction bands ($s$-type atomic orbitals, $l = m = 0$). One has $u_{c\uparrow}(r, \uparrow) = u_{c\downarrow}(r, \downarrow) = \sqrt{\omega_{\alpha}(\mathbf{r})}/\sqrt{4\pi r}$. Taking into account that the orbital contributions only from the conduction bands ($s$) orbitals, $S_{\alpha}(r) = 4\varepsilon_{s}\alpha/\sqrt{3}$ is the radial part of the atomic $s$-type wave function for a given ion. Hence, using Eq. (7) transformed to spherical tensor components, the contact interaction has the matrix elements

\[ A^{(1)}_{c,\lambda\lambda} = \frac{2\varepsilon_{s}^{3}}{3\hbar} \left( S_{\lambda}^{(1)} \right)_{s_{\lambda}s_{\lambda}}, \]

where $s_{\lambda}$ is the spin projection of the electrons in band $\lambda$. From the Wigner-Eckart theorem one finds

\[ \left( S_{\lambda}^{(1)} \right)_{s_{\lambda}s_{\lambda}}' = \frac{\sqrt{3}\hbar}{2} \left\{ \frac{1}{2}\left[ 1, s_{\lambda}; \frac{1}{2}, 1; \frac{1}{2}, s_{\lambda}' \right] \right\}. \]

Hence, the non-zero matrix elements of the spherical components of the spin operator are

\[ \left( S_{\lambda}^{(1)} \right)_{\uparrow\uparrow} = -\left( S_{\lambda}^{(1)} \right)_{\downarrow\downarrow} = 1, \]

\[ \left( S_{\lambda}^{(1)} \right)_{\uparrow\downarrow} = \left( S_{\lambda}^{(1)} \right)_{\downarrow\uparrow} = \frac{1}{\sqrt{2}}. \]

Collecting the elements of the $6c6c$ block and converting to Cartesian components one finds $A_{\alpha\beta} = \xi_{s}^{3}a_{\alpha}(\mathbf{r})|2\sigma_{\epsilon}/3$, hence $H_{6c6c} = 3A_{c}\cdot I/\hbar = \sigma\cdot I/(\hbar|a_{\alpha}(\mathbf{r})|2)$, which proves Eq. (12a).

\[ \text{b. The orbital term of the dipole part} \]

For the local term of $A_{c}$ (the second term in Eq. (2)) one substitutes the decomposition in Eq. (9) into Eq. (6).

The hydrogen-like orbitals $f_{lm}(\mathbf{r})$ building the Bloch function according to Eq. (9) are decomposed into their radial parts $R(r)$ and angular parts described by spherical harmonics $Y_{l,m}(\Omega)$. Taking into account that the components of the angular momentum operator are diagonal in spin $s$ and in the total angular momentum $l$, one gets

\[ A^{(1)}_{\alpha\beta\lambda\lambda} = \xi_{s}^{3} \sum_{lmm'x} M_{l0}^{(s)} c_{lm}^{(s)} c_{lm'}^{(s)} \langle lm'|L_{q}^{(1)}|lm \rangle, \]

where

\[ M_{l0} = \frac{a_{\beta}^{3}}{4\varepsilon_{s}} \int \text{d}r r^{2} R_{l}^{*}(\mathbf{r}) \frac{1}{r^{3}} R_{l}(\mathbf{r}). \]

Following Ref. 6 we denote $M_{11} = M_{p}$, $M_{22} = M_{d}$, $M_{02} = M_{20} = M_{ad}$. The matrix elements $\langle lm'|L_{q}^{(1)}|lm \rangle$ can be trivially calculated by elementary methods. However, a more compact and uniform result is obtained via Wigner-Eckart theorem,

\[ \langle lm'|L_{q}^{(1)}|lm \rangle = \frac{\langle l, 1; m, q|l, 1; m \rangle}{\sqrt{2l+1}}. \]

The reduced matrix element is found by inspection of the component $q = 0, m = m'$ where $\langle l, 1; m, 0|l, 1; m \rangle = m/\sqrt{2l+1}$ and obviously $\langle lm|L_{q}^{(1)}|lm \rangle = m$, hence

\[ \langle l|L_{q}^{(1)}|l \rangle = \sqrt{l(l+1)(2l+1)}. \]

Hence, the final formula is

\[ \langle \lambda|A_{c,SR,q}\lambda \rangle = \frac{\langle \lambda|L_{q}^{(1)}|\lambda \rangle}{(\hbar\alpha_{\lambda}^{(X)^*}a_{\lambda})^{2}} = \xi_{s}^{3} \sum_{lmm'x} M_{l0}^{(s)} c_{lm}^{(s)} c_{lm'}^{(s)} \sqrt{2l+1}(l, 1; m, q|l, 1; l, m'). \]

We note that this matrix element is diagonal in $l$ and vanishes for $l = 0$, hence non-zero matrix elements appear only within the valence band. Moreover, for the heavy-hole (hh) bands, in the simple single-band approximation, the Bloch functions are spin eigenstates with opposite spin orientation. Since the orbital contribution is spin-diagonal, in the single-band, purely $p$-wave model of the hh band, this term yields only a diagonal (Ising) coupling. This coupling (hence the value of the Overhauser field) is affected by band mixing only in the second order, since neither the spin-down nor the $m' = 0$ spin-up admixture couple to the leading-order ($m = 1$ spin-up) component of the nominally spin-up hh state via the $q = 0$ tensor component (due to spin conservation and $m + q = m'$ selection rule, respectively). A $d$-shell admixture introduces a $l = 2, m' = -1$ spin-up correction to the spin-up hh state (see the explicit compositions of the Bloch states in Ref. 6). This is not coupled to the leading-order ($l = 1$) component of this state but couples to the same ($l = 2, m = -1$) admixture, leading to a correction to the Overhauser field in the quadratic order in $\alpha_{d}$.

In addition, with band mixing, the nominally $+3/2$ (spin-up) hh state ($m' = 1$) may attain an admixture of the spin-down light hole state with $m' = 0$. According to Eq. (A2), this admixture is coupled to the dominating component of the $-3/2$ (spin down) hh state ($m = -1$) via the $q = 1$ component of the hyperfine coupling, thus leading to the appearance of spin flip-flop terms in the Hamiltonian. The $d$-wave admixture to hh Bloch functions are spin-conserving, hence they can only lead to spin flip-flops in combination with band mixing.
c. The spin term of the dipole part

The third term in Eq. (2) can be written in terms of cartesian components as

$$A_{s,i} = \sum_j T_{ij} S_j / \hbar,$$

where

$$T_{ij} = \frac{a_B^3}{4} 3x_i x_j - r^2 \delta_{ij} / r^3$$

is a traceless, symmetric, second order Cartesian tensor, hence its components form a second order spherical tensor. The spherical components of $A_{s}$ are

$$A_{s,q}^{(1)} = -\sqrt{15} \sum_{q_1, q_2} (2; 1, q_1, q_2 | 2; 1, q) T_{q_1, q_2}^{(2)} S_{q_2}^{(1)},$$

where the spherical components of $T_{q}^{(2)}$ are constructed from the first order position tensor $r^{(1)}$ according to the tensor multiplication rule,

$$T_{q}^{(2)} = \frac{a_B^3}{4 \pi} \sum_{q_1, q_2} \langle 1, 1, q_1, q_2 | 1, 2, q \rangle r_{q_1, q_2}^{(1)} r_{q_2}^{(1)},$$

and the overall factor has been determined by inspection.

The matrix element of $T_{q}^{(2)}$ between two hydrogen-like orbitals is

$$\int d^3r \hat{f}^{*}_{im}(r) T_{q}^{(2)} f_{lm}(r) = \sqrt{\frac{8 \pi}{15}} M_{q}; G_{q_{1}q_{2}}^{m' m''},$$

where $G_{q_{1}q_{2}}^{m' m''}$ are Gaunt coefficients,

$$G_{q_{1}q_{2}}^{m' m''} = \int d\Omega Y_{l_{1}m_{1}}^{*}(\Omega) Y_{l_{2}m_{2}}^{*}(\Omega) Y_{l_{3}m_{3}} m_{3}'(\Omega)$$

$$= (-1)^{m'} \sqrt{\frac{(2l_{1} + 1)(2l_{2} + 1)(2l_{3} + 1)}{4 \pi}}$$

$$\times \begin{pmatrix} l & l' & l'' \\ 0 & 0 & 0 \\ m & m' & m'' \end{pmatrix},$$

and $\begin{pmatrix} l & l' & l'' \\ m & m' & m'' \end{pmatrix}$ are Wigner 3-j symbols. From the parity rule on the Gaunt coefficients, $l + l' + l''$ - even, and the triangle rule, $|l - l''| \leq l' \leq l + l''$, the only non-zero contributions in Eq. (A3) are those with $(l, l') = (0, 2), (2, 0), (1, 1), (2, 2)$. Hence, non-zero matrix elements appear within the valence band and between the valence and conduction bands.

Upon substituting the result from Eq. (A3), along with the decomposition in Eq. (9), to Eq. (7) one gets

$$\frac{\langle \lambda' | A_{s,SH,d}^{(1)} \rangle}{(\hbar a_{\alpha}^{(\lambda)} a_{\alpha}^{(\lambda)})} = -\sqrt{15} \sum_{lms} \sum_{l'm's'} M_{l'l'}$$

$$\times \sum_{q_{1}, q_{2}} (2; 1, q_{1}, q_{2} | 2; 1, q) G_{l_{1}l_{2}l_{3}}^{m_{1}m_{2}m_{3}} \langle \lambda_{s}^{(1)} \rangle_{s_{l}s_{l}'} S_{q_{2}}^{(1)}.$$ 

The structure of this term is much more complicated than that of the orbital contribution, since the present term is not diagonal in $l$ and $s$. The Clebsch-gordan coefficients require $q_{1} + q_{2} = q$, while the Gaunt and $M_{l'l'}$ coefficients impose the selection rules $m' = m + q_{1}$ and $(l', l) = (1, 1), (2, 2), (0, 2),$ or $(2, 0)$.

We start with analyzing the corrections to the heavy-hole Overhauser field (Ising term, $q = 0$). The three non-vanishing decompositions are now $q_{1} = \pm 1$, $q_{2} = \mp 1$ and $q_{1} = 0$, $q_{2} = 0$. The contribution $(l', l) = (1, 1)$ yields the leading-order (hh-hh) part of the Ising coupling ($q_{1} = q_{2} = 0$) as well as coupling between components that differ by spin-orbital angular momentum flip-flop (e.g., $m' = 1$, $s' = \mp$ to $m = 0$, $s' = \pm$). For the $p$-wave component, the latter is only possible for light hole states, hence the resulting correction must rely on light-hole admixtures at both hh states and is therefore quadratic in band-mixing amplitudes. The contribution $(l', l) = (2, 2)$ clearly involves $d$-wave contributions to both states and is therefore always quadratic in the $d$-wave amplitude $\alpha_d$. It contains the contribution from the leading-order component of the hh state as well as the spin-orbital flip-flop couplings between light-hole components of the hh state, which are, additionally, quadratic in band mixing. The term with $(l', l) = (0, 2)$ couples the conduction band (cb) admixture to the $d$-wave component of the leading-order contribution of the hh state. It is, therefore, linear in $\alpha_d$ but one should remember that the cb admixture is very small.

In addition, band mixing and $d$-wave contributions to Bloch functions generate terms with $q = \pm 1$ in the Hamiltonian, that is, flip-flop couplings between the hh states and the nuclei. For instance, for $(l', l) = (1, 1)$, there is a contribution from the $q_{1} = 0$, $q_{2} = 1$ term, coupling the leading-order contribution to the hh state $(m = 1$, spin up) with a light-hole admixture to the other hh state $(m = 1$, spin down), which is linear in band mixing and therefore should be much larger than the band-mixing corrections to the Overhauser term. Another such coupling appears for $(l', l) = (2, 2)$ and $q_{1} = 2$, $q_{2} = -1$. This one couples the $m = 1$ spin-down and $m = -1$ spin-up components, that is, the $d$-wave components of the leading contribution to the two opposite hh states.

Eq. (A2) together with Eq. (A4), upon converting to Cartesian components and explicit evaluation, yield the matrix representation used in Eqs. (12b)–(12f).
