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Anisotropy of dipolar-broadened nuclear spin resonance

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Abstract. We study anisotropy of the homogeneously broadened nuclear spin resonance in a crystal. Using a combinatorial method to restrict the nuclear states to a fixed polarisation, we obtain the first four cumulants of the full line-shape as a function of polarisation of the nuclei and angles between the external magnetic field and the crystalline axes. We demonstrate that the line width is relatively isotropic but the other cumulants such as line shift and higher then the second cumulants are strongly anisotropic. Also the angular dependencies of the first three cumulants are found to be independent of the polarisation but the fourth cumulant changes its angular dependence with a change of the nuclear polarisation. This result shows that the orientation of the external magnetic field is an essential parameter in a study of polarised nuclei.

1. Introduction
Recent interest in studying nuclear spins was fuelled [1, 2, 3, 4, 5, 6, 7, 8] by the dephasing effect in GaAs quantum dots which is induced by fluctuations of the nuclear (Overhauser) field and blocks prospective application of electron spins for spintronics [9] and quantum computing [10]. Among different approaches to this problem, there is a new tool that combines radio frequency excitation and dynamical nuclear pumping techniques to control the Overhauser field and simultaneously to detect its state [11]. In particular, this method allows to access directly some of the local properties of the nuclear bath.

At infinite temperature, a connection between the line width of the nuclear magnetic resonance and the dipole interaction between nuclei in a crystal was established in [12]. Later, it was found that the line shape changes at low spin temperatures [13]. A detailed understanding of the effect of a non-equilibrium polarisation, as occurs in dynamical pumping experiments, may give a way to measure directly the degree of polarisation reached under dynamical pumping and provide further insights in the effect of dipole interaction which plays an important role in the intrinsic dynamics of the nuclear bath in unstrained structures.

In present paper we address this problem by analysing the long wavelength spectrum of polarised nuclei using a cumulant expansion of the full line shape. We employ a combinatorial method to restrict the nuclear states to a fixed polarisation and obtain the first four moments of the full line shape as a function of the nuclear polarisation and angles between the external magnetic field and axes of a crystal. We find that the polarisation and the angular dependencies...
factorise for the first three moments but are mixed for the fourth moment. For a Zince-Blende lattice of GaAs we find that the angular part of the second cumulant is almost a constant but the other cumulants change by an order of magnitude as the angles change. This result shows that the orientation of the magnetic field is an essential parameter when one wants to study the dipolar interaction between nuclei at a finite polarisation.

The paper is organised as follows. In Section II, we approximate the model of dipole interaction between nuclear spins for a large external magnetic field. In Section III, we analyse the shape the absorption line to obtain its polarisation and angular dependencies. In Section IV, the angular dependencies of the cumulants are studied for a crystal with the Zinc-Blende symmetries.

2. Dipole interaction

The Hamiltonian that describes $N$ nuclear spins located at the sites of a regular lattice, their dipole interaction with the strength $g$, and an external magnetic field $B$ is given by

$$H = \mu B \sum_j I_j^z + g \sum_{i<j} \left( 3 (e_{ij} \cdot I_i) (e_{ij} \cdot I_j) - I_i \cdot I_j \right) / r_{ij}^3,$$

where $\mu$ is the nuclear magneton, $I_j^± = I_j^z \pm i I_j^y$ are the spin-1/2 operators, index $j$ labels the sites on the lattice, $r_{ij}$ is the distance between two sites, and $e_{ij} = r_{ij}/r_{ij}$ is the unit vector that connects sites $i$ and $j$. We consider a $d$-dimensional crystal with $N = L^d$ nuclear spins and assume periodic boundary conditions, $I_{j+L} = I_j$, to exclude edge states from the analysis.

In a strong magnetic field the Hilbert space is partitioned by a large Zeeman energy into a set of subspaces labelled by the total $z$-projection of all spins, $S_z$ is an eigenvalue of $\sum_j I_j^z$. In each subspace the many spin states that have the same Zeeman energy are split by a part of the dipole interaction that conserves $S_z$,

$$H_0 = \mu B \sum_j I_j^z + \sum_{i<j} g \frac{3 \gamma_{ij}^2 - 1}{r_{ij}^3} \left( I_j^z I_i^z - \frac{I_j^+ I_i^-}{2} \right).$$

The matrix elements of the $S_z$ non-conserving remainder, $H_1 = \sum_{ij} 3g \left[ (\alpha_{ij} + i \beta_{ij})^2 I_i^+ I_i^- + 4 (\alpha_{ij} + i \beta_{ij}) \gamma_{ij} I_i^+ I_i^- + (\alpha_{ij} - i \beta_{ij})^2 I_i^- I_i^+ + 4 (\alpha_{ij} - i \beta_{ij}) \gamma_{ij} I_i^- I_i^+ \right] / 8r_{ij}^3$, connect subspaces with different Zeeman energies and can be treated perturbatively when the magnetic field is large. Here $(\alpha_{ij}, \beta_{ij}, \gamma_{ij})$ are coordinates of the vector $e_{ij}$ in a reference frame with $z$-axis parallel to the external $B$-field.

3. Absorption line shape and cumulant expansion

The absorption of electromagnetic radiation in a strong magnetic field is dominated by a flip of a single nuclear spin at an energy of $\mu B$, neglecting the dipole interaction. The dipole interaction of a single spin with other nuclear spins on the lattice makes the width of the resonance finite. Following [12] we study moments of this line shape given by the transition energies $E_f - E_f'$ between pairs of the eigenstates $|f\rangle$ and $|f'\rangle$,

$$\langle m^k \rangle = \frac{\sum_{f'f} (E_f' - E_f)^k |\langle f' | I^+ | f \rangle|^2}{\sum_{f'f} |\langle f' | I^+ | f \rangle|^2},$$

where $I^+ = \sum_j I_j^+$ is the operator that corresponds to a long wave-length photon and $k$ is the order of the moment. To account for a finite polarisation, the initial states $f$ are restricted to a subspace of fixed $S_z$. In our study of the dipolar broadening we will use only the truncated part $E_f |f\rangle = H_0 |f\rangle$ of the dipole Hamiltonian $H$. A perturbative treatment of $H_1$ produces extra peaks at $2\mu B$ and $3\mu B$ with small amplitudes $\sim g/B$, which are neglected.
The sums in Eq. (2) can be transformed to traces by means of general quantum mechanical identities. In the denominator, the sum over the eigenstates $|f\rangle$ gives a unit matrix, $\sum_f |f\rangle\langle f| = \hat{1}$, in the subspace of $S^z + 1$ as the operator $I^z$ changes $S_z$ by 1 only. Then, the remaining sum over a complete set of the eigenstates is $\sum_f \langle f | I^- | f \rangle = \text{Tr}(I^- I^+)$ where the trace is restricted to a subspace of fixed $S^z$. Representing the eigenenergies as $E_f = \langle f | H_0 | f \rangle$ the numerator can be transformed in a similar way. Explicit expressions for the first three moments are

$$
\langle m \rangle = \frac{-\text{Tr}([H_0, I^-] I^+)}{\text{Tr}(I^- I^+)} \quad , \quad \langle m^2 \rangle = \frac{-\text{Tr}([H_0, I^-] [H_0, I^+] )}{\text{Tr}(I^- I^+)}
$$

(3)

$$
\langle m^3 \rangle = \frac{\text{Tr}([H_0, [H_0, I^-]] H_0, I^+)]}{\text{Tr}(I^- I^+)} 
\langle m^4 \rangle = \frac{\text{Tr}([H_0, [H_0, I^-]] [H_0, [H_0, I^+] ])}{\text{Tr}(I^- I^+)}
$$

(4)

where $[A, B] = AB - BA$ is the operator commutator. Note, for calculating the absorption spectrum, that $I^+$ has to appear on the right of $I^-$ in the traces.

Unlike the methods of exact diagonalisation [14, 15], in this paper we will use the approach of [12] to evaluate the quantum mechanical averages. As a trace is invariant with respect to a change of basis we will calculate Eq. (3) in the basis of non-interacting spins that are quantised individually along the external magnetic field. In the basis of non interacting states, the denominator is a single sum of traces of individual spin operators $\sum_i \text{Tr}(I^-_i I^+_i) = \sum_i \text{Tr}(I^-_i I^+_i)$. The full set of the basis states in a subspace of fixed $S^z$ can be divided in two groups: one has the $i^{th}$ spin in the state $I^z_i = 1/2$ and the other in the state $I^z_i = -1/2$. For each state form the first group the expectation value of $I^-_i I^+_i$ is 0 and for the second group it is 1. Then, the trace of every single spin operator is independent of $i$, $\text{Tr}(I^-_i I^+_i) = 0 \cdot C^N_{N-1} + 1 \cdot C^N_{N-1}$, where the binomial factors $C^N_{N-1}$ and $C^N_{N-1}$ give the numbers of states in each group. Here $n$ is the number of two-spins pointing up with respect to the fully polarised state, i.e. $S^z = -N/2 + n$. The sum over all $N$ single spin traces gives the denominator as $\text{Tr}(I^- I^+) = N!/(n!(N-n-1)!)$.

The numerator of the first moment $k = 1$ requires only one first order commutator, $[H_0, I^-] = -\mu B \sum_{j=1}^N I^z_j - \sum_{i<j} \frac{1}{r_{ij}^3} 3 (\gamma_j^z - 1) I^z_i I^z_j$. The trace of a two spin operator, that is involved in Eq. (3), is evaluated analogously to the denominator. We calculate averages over
all possible two spin states accounting for the remaining \( N - 2 \) spin states by binomial factors,

\[
\text{Tr} \left( I_j^z I_i^- I_i^+ \right) = \frac{(1 - \delta_{ij}) (N - 2)! (N - 2n - 1)!}{2n! (N - n - 1)!}.
\]

Substituting the commutator \([H_0, I^-]\) into Eq. (3) and using the last expression we obtain the line shift, \( \langle \nu \rangle = \langle m \rangle \), that corresponds to the first moment \( \langle \nu \rangle = \mu B - F^3 (N - 2n - 1) / (2N - 2) \). Evaluating the limit \( N \to \infty \) of this result and identifying the polarisation as \( p = -1 + 2n/N \) we obtain the expression in Table. 1. The first moment of the crystalline field \( F \) is given in the same table.

The numerator of the second moment \( k = 2 \) requires an extra first order commutator

\[
[H_0, I^+] = B \sum_j I_j^+ + \sum_{i<j} \frac{2}{r_{ij}^2} 3 \left( 3z_i^2 - 1 \right) I_i^+ I_i^z.
\]

The trace of a three spin operator, which is needed for \( \langle m^2 \rangle \), is obtained in a similar way,

\[
\text{Tr} \left( I_j^z I_i^- I_i^+ I_i^z \right) = \frac{(1 - \delta_{ij}) (1 - \delta_{ii'}) (N - 3)! (2n - N + 1)(2n - N + 2) - \delta_{ii'} 2n(2n - 2N + 3))}{4n! (N - n - 1)!}.
\]

Substituting the product of the two commutators \([H, I^-] [H, I^+]\) into Eq. (3), and using the above expression, we obtain the second cumulant \( \langle \nu^2 \rangle = \langle m^2 \rangle - \langle m \rangle^2 \) that corresponds to the line width, \( \langle \nu^2 \rangle = F_2 3n (2N - 2n - 3) / (2 (N - 1) (N - 2)) \). Evaluating the \( N \to \infty \) limit of this expression we obtain the result in Table. 1.

The third and the fourth moments contain an admixture of moments of a lower order when the polarisation is finite, \( p \neq 0 \). Hence, we evaluate the third and the fourth irreducible moments as

\[
\langle \nu^3 \rangle = \langle m^3 \rangle - 3 \langle \nu^2 \rangle \langle m \rangle - \langle \nu \rangle^3 \quad \text{and} \quad \langle \nu^4 \rangle = \langle m^4 \rangle - 4 \langle \nu^3 \rangle \langle m \rangle - 3 \langle \nu^2 \rangle^2 - 6 \langle \nu \rangle^2 \langle \nu^2 \rangle - \langle \nu \rangle^4,
\]

using the same procedure as for the moments above. In the limit \( N \gg 1 \), we obtain the results in Table. 1. The dependencies of \( \langle \nu^k \rangle \) on the polarisation \( p \) and the moments of the crystalline field \( F_1 \) factorise for \( k \leq 3 \). This in turn implies that the dependence of the model Eq. (1) on the orientation of the external magnetic field manifests itself equivalently in spectroscopy at different polarisations of the nuclei. However, for \( k = 4 \) such factorisation does not occur thus the anisotropy changes with a change of the polarisation.

4. Crystalline anisotropy

The moments of the crystalline field \( F_1 \) depend on the orientation of the external magnetic field. As an example we evaluate the prefactors in Table. 1 numerically for the zinc-blende crystals; GaAs belongs to this class.

These crystals contain two species of atoms. In a typical spectroscopy experiment [4] all nuclei are polarised first then they are probed at a selected frequency to get a signal from a single sublattice while the other sublattice, at a different frequency, stays polarised that makes contribution of the other sublattice small [16]. Having in mind such a setup we will study only one sub-lattice.

The angular dependence of the second moment \( \sqrt{F_2} \) is presented in Figure 1b. The shape is relatively isotropic, the biggest change in magnitude of \( \sqrt{F_2} \) is \( \sim 20\% \) between the directions [001] and [111]. The remaining cumulants are strongly anisotropic. The first moment is the biggest along the direction [111] and in the plane (111), see Figure 1a. In all other directions it is an order of magnitude smaller. The third cumulant is maximum along some twelve directions and is zero along some other twelve directions with a smooth cross-over in-between, see Figure. 1c. The angular dependence of the fourth cumulant at \( p = 0 \) is present in Figure 1d and the second part of the cumulant that becomes non-zero at \( p \neq 0 \) is plotted in Figure 4e.
Figure 1. Plots of a) $F$, b) $\sqrt{F^2}$, c) $\sqrt{F_3c - F_2F - F_4}$, d) $F_{4y} - 2F_4 - F_2^2$, and e) $F^2F_2 + 10FF_3 - 2FF_3c + F_{4c}$ as a function of angles between B and the axes of a Zince-Blende crystal.

5. Conclusions
We have studied anisotropy of the homogeneously broadened nuclear spin resonance at a finite polarisation in a crystal with respect to mutual orientation of the external magnetic field and axis of the crystal. We have demonstrate that the line width is relatively isotropic but the other cumulants, line shift and higher then the second cumulants, are strongly anisotropic. This result shows that the orientation of the magnetic field is an essential parameter in a study of polarised nuclei.

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