Influence of a stray magnetic field on the measurement of long-range spin-spin interaction

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Abstract
We study the influence of an additional uncontrolled (stray) magnetic field upon the measurement of long-range spin-spin interaction strength of two spin-1/2 valence electrons bound in two separate ions at well-defined distances from each other. This stray field, which is neither perpendicular nor parallel to the line connecting two ions, could appear due to the Earth magnetic field, or, due to the slight angular misalignment between the applied magnetic field and the line connecting two ions. It is found that the presence of the stray magnetic field plays an important role in the dynamics of the spin-states of two electrons. If neglected in the analysis, moreover, such a stray field may affect the measurement of the spin-spin interaction strength, especially at smaller inter-spin distances.

1. Introduction

The spin-spin interaction is one of the basic interaction of electrons that arises from the coupling between the internal spin-magnetic moments of two electrons. In atomic physics this interaction of spins is usually treated together with other relativistic effects within the Breit operator [1]. The magnetic interaction has been tested to a very high accuracy, e.g., in the ground-state energy of heliumlike uranium ion [2, 3]. In some cases, moreover, it could even dominate the Coulomb repulsion [4], e.g., the angular distribution of x-ray emission following the dielectronic recombination changes drastically due to the interaction of magnetic moments [5, 6]. Although the spin-spin interaction is known to be important for predicting accurate atomic and molecular energies, on the long-range scale it rapidly decreases as an inverse-cube power with the distance among the electrons. This rapid decrease makes the observation of this spin-coupling rather difficult. However, the interaction among two electron spins has been investigated recently in several experiments, for instance with nitrogen-vacancy (NV) defects in diamond [7–10], dopant atoms in semiconductors [11], single-molecule magnets in break junctions [12], bound electrons in two separated ions [13], as well as atoms in a scanning tunneling microscope [14]. Further interest in these spin-spin interactions arises from the applications in spintronics and quantum computation [15], electron spin resonance spectroscopy [16], as well as in the search for a new physics [17].

Here, special emphasis has been placed upon the experiments by Kotler et al [13], in which the long-range spin-spin interaction strength between bound electrons has been accurately measured at distances of several micrometers. In particular, the spin-spin interaction between two 5s1/2 valence electrons bound in two separate 88Sr+ ions has been investigated. In the experiments by Kotler et al [13], two ions were placed into a linear Paul trap, and with a magnetic field parallel to the trap axis. In such a set-up, the interaction strength can be extracted from the magnetic-field-independent spin-spin evolution within the subspace that is spanned by the two Bell states. Due to the applied magnetic-field insensitivity, it became possible to measure very weak (millihertz-scale) spin-spin interaction strength in the presence of a magnetic noise that was six orders of magnitude larger than the magnetic fields of the electrons acting on each other. Initially, the electron spins were prepared in the state. This was achieved by the micro-motion sideband excitation placing one of the ions at the radio frequency null inside the trap while another one at 2–3 μm away from the null. Under the spin-spin interaction initially prepared state evolved in time within the decoherence-free subspace spanned by states. Such an...
evolution can be understood as a rotation with a rate proportional to the spin-spin interaction strength, which was deduced from the experiment to be $2\pi \times 1.1(2)$ mHz for the case of $2.4 \mu m$ inter-ion distance [13]. Although the theoretical value $2\pi \times 0.93$ mHz lies within the given experimental error bars, it differs by about 18%. To understand such deviations between the measured and theoretically estimated values, we here analyze the effect of an uncontrolled (stray) magnetic field on the measurement of the long-range spin-spin interaction strength of electrons.

In this paper, we shall work in SI units and keep all factors of $\hbar$ and $c$ in the formulas to directly relate our estimates and discussions to the experiments with ion traps or nitrogen centers in diamonds. Indeed, the use of SI units appears to be useful for analyzing the spin-dependent interactions at a separation of the spin between $\mu m$ to nm. The paper is organized in the following way: section 2 describes the theoretical background of the calculations. This includes the explanation of different components of the magnetic fields in the two electron system in section 2.1, the spin-dependent Hamiltonian of electrons in section 2.2, and the scheme of measurement of the spin-spin interaction strength in section 2.3. In section 3, we then present and discuss our results. Finally, a short summary and outlook are provided in section 4.

2. Theoretical background

In this article, we present and discuss the influence of an additional unaccounted magnetic field on the description of the measurement of long-range spin-spin interaction strength between electrons. Such a stray field could originate, e.g., due to the slight angular misalignment between the external magnetic field and the line connecting two ions or due to the Earth magnetic field. In order to understand the role of this effect on the measurement of long-range spin-spin interaction, let us first explore how a magnetic field colinear and perpendicular to the field affects the time-evolution of an initially prepared spin state.

2.1. The components of magnetic fields in the system

In the measurement [13] it was assumed that in the system there is only an applied magnetic field $\vec{B} = \vec{B}_{tot}$, which is directed along the line connecting the two ions, cf figure 1(a). Here, we consider the presence of an additional (stray) magnetic field $\vec{B}_s$ in the system. Without loss of generality one can assume that it acts along the perpendicular direction to the line connecting the two ions, since the other component of the stray magnetic field can be added to the applied field. Hence, the total magnetic field $\vec{B}_{tot}$ can be expressed as follows

$$\vec{B}_{tot} = \vec{B}_s \hat{n} + \vec{B}_p,$$

where $\hat{n}$ and $\vec{p}$ represent the two unit vectors, one is perpendicular and another is parallel to the line connecting the two ions, cf figure 1(b). We, particularly, consider these two unit vectors laying in the $xz$ plane are perpendicular to each other. In order to illustrate the possible role of the stray magnetic field in the measurement of the spin-spin interaction, we have drawn the schematics of the two-electron system in figure 1, where the initially prepared state $|\uparrow \downarrow\rangle$ is sketched for both of the cases. Despite the spin-spin interaction does not depend on an applied magnetic field, the measurement outcome depend on spin basis functions employed for its detection. While the spin basis functions are the spin states with defined projection of the spin momentum on a
direction of total magnetic field. Thus, if the perpendicular field component \( B_{\perp} \) is acting in the system, the total magnetic field \( \vec{B}_{\text{tot}} \) is the vector sum of \( \vec{B} \) and \( \vec{B} \) with \( B_{\text{tot}} = \sqrt{B_1^2 + B_2^2} \). Therefore, an angle \( \theta \) is created between \( \vec{B}_{\text{tot}} \) and the line connecting two ions as shown in figure 1(b). In present work we aim to investigate the effects due to the presence of a stray magnetic field in the measurement of long-range spin-spin interaction strength.

2.2. Spin-dependent Hamiltonian

In order to understand the effect of the perpendicular field component on the long-range spin-spin interaction measurement, we start with the spin-part of the two-electron Hamiltonian. The spin-spin interaction operator of two electrons can be expressed with the help of the Breit operator [1], as

\[
V_{SS} = \frac{\mu_B}{4\pi} \sum_{i<j} \frac{\sigma_i \cdot \sigma_j}{|\vec{r}_{ij}|^3} - \frac{3(\sigma_i \cdot \vec{r}_{ij})(\sigma_j \cdot \vec{r}_{ij})}{|\vec{r}_{ij}|^5},
\]

Here, \( \mu_B \) is the vacuum permeability, \( \mu_B \) is the Bohr magneton (\( \mu_B = e\hbar/2m \)), and \( \sigma_i \) is the Pauli spin matrix acting on \( i \)th electron. In equation (2), \( \vec{r}_{ij} \) describes the separation distance between two electrons. The electrons are considered to be bound in two separate ions, which are located at well-defined distance \( d \) from each other. Since the distance between ions is much larger than a typical atomic size, i.e., \( d \gg a_0 \), where \( a_0 \) is the Bohr radius, one can use the point-dipole approximation, \( |\vec{r}_{ij}| \approx d \). Thus, the spin-spin interaction operator of two electrons is approximated by

\[
V_{SS}^{\text{dep}} = \frac{\mu_B}{4\pi} \sum_{i<j} \frac{\sigma_i \cdot \sigma_j}{d^3} - \frac{3(\sigma_i \cdot \vec{d})(\sigma_j \cdot \vec{d})}{d^5}.
\]

Here, \( \vec{d} = (\xi \cos \theta - \xi \sin \theta) \) represents the inter-ion distance in a chosen coordinate system. The vector \( \vec{d} \) forms an angle \( \theta \), \( \theta = \tan^{-1}(B_{\perp}/B) \) with the spin-quantization axis \( z \). Since within the employed approximation the spin-spin interaction operator does not depend on spatial coordinates, the electron spin dynamics can be described by a reduced spin Hamiltonian (\( H_{\text{spin}} \)), which acts only on spin coordinates of the electron wave functions. In addition to the spin-spin interaction one has to include the interaction of the spins with the total magnetic field \( \vec{B}_{\text{tot}} \). As a result the spin Hamiltonian takes a form:

\[
H_{\text{spin}} = \mu_B B_{\text{tot}} (\sigma_1 \cdot \sigma_2) + \hbar \xi (2\sigma_1 \sigma_2 - \sigma_1 \sigma_2 - \sigma_1 \sigma_2) - \hbar \Delta (\sigma_1 \sigma_2 - \sigma_1 \sigma_2),
\]

where \( B_{\text{tot}} \) represents the magnitude of the total magnetic field, which direction is chosen as \( z \) axis, \( \xi = \mu_B \mu_0^2/(4\pi \hbar) \times 1/d^3 \) defines the spin-spin interaction strength between the electrons, while \( \Delta \) and \( \Delta \) describe the corrections originated due to the misalignment of the quantization axis and the line connecting two ions, \( \Delta \) \( \Delta \) are zero and the last two terms vanish, thus, the Hamiltonian used in [13] is restored. The terms \( \sigma_1 \sigma_2 \) can be understood as a shift of the spin-flip energy of one of the electron conditioned on the spin-state of another electron. The terms \( \sigma_1 \sigma_2 \) and \( \sigma_1 \sigma_2 \) lead to the collective spin-flips in which a spin excitation is interchanged, while the terms proportional to \( \Delta \) result in the single spin-flips. Here, we note that single spin-flips are only allowed when \( \theta = 0 \). The spin Hamiltonian can be also represented in the matrix form, e.g., in the basis of spin functions \( |\uparrow\rangle, |\downarrow\rangle \), \( |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle \), it is given by:

\[
H_{\text{spin}} = \hbar \begin{pmatrix}
2\xi - \delta_1 + \omega_z & -\delta_1 & -\delta_2 & 0 \\
-\delta_1 & -2\xi + \delta_1 & -2\xi + \delta_1 & \delta_2 \\
-\delta_2 & -2\xi + \delta_1 & -2\xi + \delta_1 & \delta_2 \\
0 & \delta_2 & \delta_2 & 2\xi - \delta_1 - \omega_z
\end{pmatrix},
\]

where \( \omega_z = 2\mu_B B_{\text{tot}}/\hbar \) is the Larmor frequency of electron spin precession in the magnetic field \( \vec{B}_{\text{tot}} \). As it can be clearly seen from (5) the matrix elements proportional to \( \Delta \) mix the states which differ by the spin state of one of the electron. In order to investigate the spin evolution driven by the spin-dependent Hamiltonian matrix (5), we solve the time dependent Schrödinger equation of the two electron system

\[
i\hbar \frac{\partial}{\partial t} \psi(t) = H_{\text{spin}} \psi(t),
\]

where \( \psi(t) \) describes the spin wave function of the two electron system, which can be decomposed as follows

\[
\psi(t) = A(t) |\uparrow\rangle + B(t) |\downarrow\rangle + C(t) |\uparrow\downarrow\rangle + D(t) |\downarrow\uparrow\rangle.
\]
and subspace. This implies that the probability of having the spin-states of two electrons in the subspace is leaked to the subspace for the case of \( \theta = 0^\circ \) and the inter-ion distance \( d = 2.41 \mu m \). Therefore, it is foreseen that the measurement of the interaction strength, which is based on the analysis of dynamics of the spin-states in the \( | \uparrow \rangle \) and \( | \downarrow \rangle \) subspace, will be more affected at larger \( \theta \) or \( d \). Hence, it is expected that the measurement of the interaction strength, which is based on the analysis of dynamics of the spin-states in the \( | \uparrow \rangle \) and \( | \downarrow \rangle \) subspace, will be influenced by the presence of the stray magnetic field.

In figure 3 we show the population probabilities \( P \) of spin-state of two electrons as a function of interaction time \( t \) on the basis functions \( | \uparrow \uparrow \rangle, | \uparrow \downarrow \rangle, | \downarrow \uparrow \rangle, \) and \( | \downarrow \downarrow \rangle \) for the case of \( \theta = 10^\circ \) and for the inter-ion distance \( d = 2.41 \mu m \). From figure 3, it can be seen that only in the case of \( \theta = 10^\circ \) the population probabilities \( P \) of spin-state of two electrons are limited to the \( | \uparrow \uparrow \rangle \) and \( | \downarrow \downarrow \rangle \) subspace at different \( t \). This implies that the probability of having the spin-states of two electrons in the \( | \uparrow \uparrow \rangle \) and \( | \downarrow \downarrow \rangle \) subspace for the case of \( \theta = 10^\circ \) is diverged for larger evolution time \( t \) from the same at \( \theta = 0^\circ \). Therefore, it is foreseen that the measurement of the interaction strength, which is based on the analysis of dynamics of the spin-states in the \( | \uparrow \rangle \) and \( | \downarrow \rangle \) subspace, will be more affected at larger \( t \) by the influence of the stray magnetic field.

2.3. Spin-spin interaction strength

When the applied magnetic field \( \vec{B} \) is perfectly directed along the line connecting the two ions, the dynamics of the spin-states of two electrons within the \( | \uparrow \downarrow \rangle \) and \( | \downarrow \uparrow \rangle \) subspace depends only on the spin-spin interaction strength \( \xi \) between the electrons. In particular, the initially prepared state \( | \uparrow \downarrow \rangle \) oscillates in time between \( | \uparrow \downarrow \rangle \) and \( | \downarrow \uparrow \rangle \) states with an oscillation rate \( 2\xi \). Thus, the spin-spin interaction strength can be accessed by measuring the visibility of such an oscillation:

\[
V = |B^*(t)C(t)|, 
\]

where \( B(t) \) and \( C(t) \) are time-dependent coefficients in the expansion of spin wave function given in equation (6). In order to measure the visibility the following experimental scheme was proposed and realized in [13]. First, the spin-state of two electrons initialized to \( | \uparrow \downarrow \rangle \) was allowed to evolve under the spin-spin interaction up to particular time \( \phi \), what creates the mixture of \( | \uparrow \downarrow \rangle \) and \( | \downarrow \uparrow \rangle \) states. Further, a controlled magnetic field gradient was applied for a fixed duration to accumulate the relative phase between these spin-states. The next step was a global \( \pi/2 \) spin rotation, which was achieved by applying an external magnetic field resonantly modulated along the direction perpendicular to the applied (constant) magnetic field \( \vec{B} \). After making the collective \( \pi/2 \) rotation, the projective measurements were performed in order to obtain the ions population at states \( | \uparrow \rangle, | \downarrow \rangle, \) and \( | \uparrow \downarrow \rangle + | \downarrow \uparrow \rangle \) by the state selective fluorescence [13]. As a result, the measured visibility in the case of absence of any perpendicular magnetic field reads

\[
A(t) = (i/2)\sin(2\delta \xi t)e^{-i\omega t}, \\
B(t) = e^{i(2\delta \xi t)}[\cos(2\xi t)\cos((2\xi t - \delta \xi t)] - i \sin(2\xi t)\sin((2\xi t - \delta \xi t)], \\
C(t) = e^{i(2\delta \xi t)}[-\sin(2\xi t)\cos((2\xi t - \delta \xi t) + i \cos(2\xi t)\sin((2\xi t - \delta \xi t)], \\
D(t) = (-i/2)\sin(2\delta \xi t)e^{i\omega t}. 
\]

\[\text{Figure 2. (a) Population probabilities } P \text{ of spin-state of two electrons for the case of } \theta = 0^\circ \text{ after an interaction time } t = 15 \text{ s and for the inter-ion distance } d = 2.41 \mu m \text{ given that the system was initialized to } | \uparrow \downarrow \rangle \text{ state. (b) The same but for the case of } \theta = 10^\circ.\]

\[\text{Figure 3. Population probabilities } P \text{ of spin-state of two electrons as the projections on the basis functions } | \uparrow \rangle, | \downarrow \rangle, | \uparrow \uparrow \rangle, \text{ and } | \downarrow \downarrow \rangle \text{ after an interaction time } t = 15 \text{ s and for the inter-ion distance } d = 2.41 \mu m. \text{ As one can see from the figure, only in the case of } \theta = 0^\circ \text{ the dynamics of the spin-states of two electrons is limited to the } | \uparrow \downarrow \rangle \text{ and } | \downarrow \uparrow \rangle \text{ subspace. But, if a stray magnetic field is present in the system, there is a nonzero probability that the spin-states of electrons can escape to the } | \uparrow \rangle \text{ and } | \downarrow \rangle \text{ subspace. This implies that the probability of having the spin-states of two electrons in the } | \uparrow \downarrow \rangle \text{ and } | \downarrow \uparrow \rangle \text{ subspace is leaked to the subspace spanned by the } | \uparrow \rangle \text{ and } | \downarrow \rangle \text{ states. Therefore, the dynamics of the spin-states within the } | \uparrow \rangle \text{ and } | \downarrow \rangle \text{ subspace becomes affected due to an angular misalignment between the magnetic field and the line joining two ions. Hence, it is expected that the measurement of the interaction strength, which is based on the analysis of dynamics of the spin-states in the } | \uparrow \rangle \text{ and } | \downarrow \rangle \text{ subspace, will be influenced by the presence of the stray magnetic field.} \]
In the case of the presence of the stray magnetic field, which is characterized by the angle \( \theta \), we repeat the experimental scheme realized in [13] for the measurement of the visibility and arrive to

\[
V_{\text{meas}}(\theta = 0) = \sin 4\xi t.
\]

(10)

In the case of the presence of the stray magnetic field, which is characterized by the angle \( \theta \), we repeat the experimental scheme realized in [13] for the measurement of the visibility and arrive to

\[
V_{\text{meas}}(\theta) = \sin(2(2\xi - \delta \xi_1)t) \pm \frac{1}{2} \sin^2(2\delta \xi_2 t) \sin(2\omega_2 t),
\]

(11)

which exactly coincides with equation (10) for the case when \( \delta \xi_1 = \delta \xi_2 = 0 \). Here, \( \pm \) sign comes from the uncertainties in the geometry of \( \pi/2 \) rotation, since the resonantly modulated external magnetic field might be applied either parallel or perpendicular to the stray magnetic field \( B_s \). Thus, the obtained results will differ exactly by the sign of the second term. Further, we will surmise that in the experiment some additional stray magnetic field is present and what is measured is the visibility given by equation (11). However, the spin-spin interaction strength is deduced by the dependence given by equation (10). Therefore, if we assume that there is a stray magnetic field, the experimentally deduced spin-spin interaction strength \( \xi_{\text{exp}} \) is given by the expression

\[
\xi_{\text{exp}} = \frac{1}{4t} \sin^{-1} \left[ \sin(2(2\xi - \delta \xi_1)t) \pm \frac{1}{2} \sin^2(2\delta \xi_2 t) \sin(2\omega_2 t) \right].
\]

(12)

where the first term corresponds to the change in the interaction strength due to the rotation of the magnetic field created by one of the electron, while the second term describes the leakage of probability to the \( \{\uparrow \downarrow \} \) subspace. In equation (12), \( \xi_{\text{exp}} \) is only an apparent interaction strength, which was measured from the oscillation rate between the \( \{\uparrow \downarrow \} \) and \( \{\downarrow \uparrow \} \) states [13]. This differs from the physical interaction strength, since that oscillation rate could depend on interaction time \( t \) by the presence of stray magnetic field as pointed out in figure 3. Equation (12) allows us to investigate how the experimentally deduced spin-spin interaction strength depends on the presence of a stray magnetic field \( (B_s) \) at different inter-ion distances \( (d) \).

### 3. Results and discussion

In this section, we study the dependence of the spin-spin interaction strength on a presence of a stray magnetic field. In figure 4, we have shown the variation in the measured spin-spin interaction strength defined by equation (12) with the magnitude of external stray field \( B_s \) as a function of inter-ion distance. It can be seen from
the figure how the measured interaction strength changes with \( d \), if the effect of the perpendicular magnetic field is taken into account. In the absence of any perpendicular magnetic field, i.e., for \( \theta = 0^\circ \), the measured spin-spin interaction strength \( (\xi_{\text{exp}}) \) does not depend on the applied magnetic field \( B \) and the interaction time \( t \), and coincides with the spin-spin interaction strength \( \xi \) given by \( \xi = \mu_B^2/(4\pi\hbar) \times 1/d^3 \). The results are calculated to be 1.252, 0.927, and 0.617 (in units of \( 2\pi \times \text{mHz} \)) for \( d = 2.18, 2.41, 2.76 \text{ \( \mu \)m} \), respectively. However, in the case of a stray magnetic field the experimental results will depend also on the applied magnetic field \( B \) and the interaction time \( t \) according to equation (12). In what follows we use the values for \( B \) and \( t \) employed in the measurement [13], i.e., \( B = 0.44 \text{ mT} \) and \( t = 15 \text{ s} \). In the case when the perpendicular magnetic field amounts to 6% of \( B \) or \( \theta = 10^\circ \), the measured spin-spin interaction strength \( \xi_{\text{exp}} \) will amount to 1.196 \( 2\pi \times \text{mHz} \), 0.885 \( 4\pi \times \text{mHz} \) for inter-ion distances 2.18, 2.41, 2.76 \( \mu \text{m} \), respectively. The values inside the brackets represent the uncertainties of the results, which originate from the second term in equation (12). In figure 4, we have plotted also the results for different values of the perpendicular magnetic field \( \theta = 20^\circ \) (36% \( B \) and \( 30^\circ \) (58% \( B \)). These particular choices of \( \theta \) are meaningful, since, in particular, a stray magnetic field can be the Earth magnetic field, which amounts to 0.044 624 mT near the experimental venue of [13]. We now compare our results with those obtained in the experiment [13] for the inter-ion distances \( d = 2.18, 2.41, 2.76 \text{ \( \mu \)m} \). It is found that the effect of the presence of a perpendicular magnetic field always leads to a decrease of \( \xi_{\text{exp}} \) \( m \text{Hz} \), and, therefore, the reduction in the interaction strength observed experimentally at \( d = 2.76 \text{ \( \mu \)m} \) might be a signature of a stray field. Overall, however, for a given magnitude of the applied magnetic field and the interaction time the influence of the stray magnetic field is rather small. For example, for the case of the stray magnetic field being of 10% of the applied magnetic field the interaction strength changes up to 2% at the distances \( d = 2–3 \text{ \( \mu \)m} \) for \( t = 15 \text{ s} \). However, for larger interaction time, for example at \( t = 30 \text{ s} \), the interaction strength \( (\xi_{\text{exp}}) \) can change up to 5% at the distances \( d = 2–3 \text{ \( \mu \)m} \) even for the case of the stray magnetic field being of 10% of the applied magnetic field. If we consider the smaller inter-ion distances the effect due to the leakage of the probability from the \( \{\uparrow \downarrow \}, \{\downarrow \uparrow \} \) subspace given by the second term in equation (12) starts to be important. This can be also seen from figure 4, where the uncertainty of \( \xi_{\text{exp}} \) strongly increases with a decrease of \( d \). Hence, for small inter-ion distances the effect of stray magnetic fields will be much more important. For that cases, the formulas and analysis presented in the current work might be especially beneficial.

4. Summary and outlook

In summary, we have investigated the role of an unaccounted (stray) magnetic field in the description of the measurement of the long-range spin-spin interaction strength between electrons. It has been shown that the dynamics of the spin-states in the \( \{\uparrow \downarrow \} \) and \( \{\downarrow \uparrow \} \) subspace becomes affected by a stray magnetic field. As a consequence, it has been found that the measurement of the spin-spin interaction strength is also influenced by the presence of this additional magnetic field. For the measurements at relatively large inter-ion distances of about 2–3 \( \mu \text{m} \) the effect is found to be of the order of several percentages. However, for smaller inter-ion distances the presence of a stray field could lead to a stronger impact. Moreover, for distances of several nm,
which is the case in the experiments with spin defects in diamonds [7], the effects beyond the magnetic-dipole approximation start to be also important [18]. This issue is currently under investigation in our group.

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