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# Spin-orbit thermal entanglement in a rare-earth-metal ion: Susceptibility witness

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In the present work, we explore the thermal entanglement between spin and orbital angular momentum in a rare-earth ion. A witness, based on the magnetic susceptibility and capable of revealing entanglement between these two angular momenta of different nature, is introduced. We found entanglement temperatures of 322 K for promethium and 715 K for samarium. These high temperatures make interesting the use of rare-earth in applications of quantum-information processes at room temperature.

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## I. INTRODUCTION

Besides purely fundamental interest [1,2], entanglement is studied as an important resource in protocols of quantum communication and information processes [2], and the main issue for the engineering of these processes is the effect of rising temperatures on the preparation of entangled states. Until a few years ago, the community thought that entanglement could not survive when the temperature rises, because, on this scale, decoherence could destroy all quantum correlations. These undesired effects usually are treated inside schemes of processing and storing quantum information that allow decoherence to be kept under control [3]. However, in some cases, the decoherence control must not be essential since several works have been demonstrated [4,5] that entangled states can be present in solids at finite and even above room temperature [6,7]; this kind of entanglement is known as "thermal entanglement."

A state of a system is said to be entangled with respect to a particular $N$ partition when it cannot be written as $\rho = \sum_i p_i \rho_i^1 \otimes \cdots \otimes \rho_i^N$, where $\sum_i p_i = 1$, $\rho_i^j$ is the density matrix of the $j$th part corresponding to the $i$th subensemble, and $p_i$ are the probabilities. This state is the most general that can be constructed using only local operations and classical communication [8]. However, in spite of the rigorous mathematical definition, characterization and quantification of entanglement is a very complex task in most cases. For bipartite pure states it is possible to characterize entanglement using entropy criteria or violations of Bell inequalities [9]. For general bipartite mixed states of arbitrary dimension there are sufficient criteria of entanglement, for example, those presented in Refs. [10,11]. But necessary and sufficient criteria only exist if we restrict the Hilbert space to $2 \otimes 2$, $2 \otimes 3$, and $\infty \otimes \infty$ dimensions [12].

A useful approach to resolve entanglement in systems of arbitrary dimensions, and one that can be useful even for multipartite systems, is the use of observables that works as entanglement witnesses. In particular, a bipartite density matrix $\rho$ is entangled if, and only if, there exists an observable $W$ that satisfies $\mathrm{Tr}\{W\rho\} \leqslant 0$ and $\mathrm{Tr}\{W\sigma\} \geqslant 0$ for all bipartite separable density matrices $\sigma$ [13,14]. The observable $W$ is usually called an entanglement witness.

In a previous work, we used an entanglement witness based on the internal energy capable of showing entanglement between spin and angular momentum in a rare-earth ion [7], and we observed entanglement in all light rare-earth. In spite of the success obtained with the energy witness, we are concerned with a method easily applicable in a laboratory. For this reason, in this work, we explore how to use the magnetic susceptibility as an entanglement witness for rare-earth ions. We are interested in thermal entanglement between effective angular momenta: spin and orbit, of a single rare-earth ion. For those who want to know about spin-orbit entanglement from a different context, there is a previous work dealing with entanglement on superexchange bonds in transition-metal oxides [15]. The interest of using rare-earth ions comes principally from their energetic configuration. The gap of energy between the ground state and the first excited state is proportional to the $S$-$L$ coupling and in all cases it is higher than room temperature. Thus, such ions are good candidates to keep entanglement up to high temperatures, an essential request for tasks that use entanglement in their performances.

## II. MAGNETIC SUSCEPTIBILITY AS ENTANGLEMENT WITNESS: SPIN-SPIN CASE

Wieśniak *et al.* [16] had shown that magnetic susceptibility can be used to reveal entanglement between spins of different ions in a solid. They considered a composite system consisting of $N$ spins with arbitrary length $s$ in a lattice, which is described by a spin Hamiltonian $H_0$. An additional term $H_1 = B\sum_{j=1}^N S_j^z$ is added when the solid is placed in a weak magnetic field of magnitude $B$, along the $z$ direction. Thus, the Hamiltonian becomes $H = H_0 + H_1$. The state of thermal equilibrium is described by $\rho = e^{-H/kT}/Z$, where $Z$ is the partition function and $k$ is the Boltzmann constant.

From the partition function, it is possible to show that if $[H_0, H_1] = 0$, the magnetic susceptibility along the $z$ axis is given by [17]

$$\chi_z = \frac{1}{kT}\left(\sum_{i,j=1}^{N}\left\langle S_i^z S_j^z\right\rangle - \left\langle \sum_{i=1}^{N} S_i^z\right\rangle^2\right), \qquad (1)$$

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TABLE I. Values of spin $s$ and orbital angular momentum $l$, as well as the energy difference $\Delta$ between the first excited state and the ground state for light rare-earth ions. The spin-orbit coupling parameter $\lambda$ is also presented [24].

| | Ce | Pr | Nd | Pm | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $s$ | 1/2 | 1 | 3/2 | 2 | 5/2 | 3 | 7/2 | 3 | 5/2 | 2 | 3/2 | 1 | 1/2 |
| $l$ | 3 | 5 | 6 | 6 | 5 | 3 | 0 | 3 | 5 | 6 | 6 | 5 | 3 |
| $\Delta$(K) | 3 150 | 3 100 | 2 750 | 2 300 | 1 450 | 500 | 43 200 | 2 900 | 4 750 | 7 500 | 9 350 | 11 950 | 14 800 |
| $\lambda$(K) | 900 | 620 | 500 | 460 | 414 | 500 | | −483 | −633 | −937 | −1 247 | −1 991 | −4 229 |

where the sum is over the spin correlations for the sites $i$ and $j$. It was shown [16] that, if the system of $N$ spins is in a separable state, the relation

$$\bar{\chi} \equiv \chi_x + \chi_y + \chi_z \geqslant \frac{Ns}{kT} \tag{2}$$

is satisfied, where $\chi_v$ is the magnetic susceptibility along the $v$ axis. Thus, the susceptibility $\bar{\chi}$ is an entanglement witness, and the proof follows below.

The question is to obtain a bound for $\langle S_i^\alpha S_j^\alpha \rangle$, where $\alpha = (x,y,z)$. For this end, let us observe that

$$\bar{\chi} = \frac{1}{kT}\left[\sum_i \left(\langle \vec{S}_i^2\rangle - \langle \vec{S}_i\rangle^2\right) + \sum_{i\neq j}(\langle \vec{S}_i\cdot\vec{S}_j\rangle - \langle\vec{S}_i\rangle\cdot\langle\vec{S}_j\rangle)\right]. \tag{3}$$

The first term in the above equation contains the variance of $\langle \vec{S}_i\rangle$, while the second contains the correlation functions. For separable states, that is, $\langle \vec{S}_i\vec{S}_j\rangle = \langle\vec{S}_i\rangle\langle\vec{S}_j\rangle$, the susceptibility (3) is reduced to

$$\bar{\chi} = \frac{1}{kT}\sum_i [\langle \vec{S}_i^2\rangle - \langle\vec{S}_i\rangle^2]. \tag{4}$$

For an arbitrary state of a spin $S$, we know the relations $\langle \vec{S}^2\rangle = S(S+1)$ and $\langle \vec{S}\rangle^2 \leqslant S^2$ hold. These expressions can be used to deduce the inequality

$$\bar{\chi} \geqslant \frac{NS}{kT}. \tag{5}$$

Note the expression above does not depend on the details of $H$; the only necessary condition is that the Hamiltonian commutes with the total magnetic moment. Placing the right-hand side of Eq. (5) on the left-hand side, we have an inequality for separable states. The violation of this inequality indicates the presence of entanglement in the state of the system. Thus, Eq. (5) defines an entanglement witness in the form

$$E_W(N) = \frac{kT}{NS}\bar{\chi} - 1. \tag{6}$$

Negative values of $E_W(N)$ mean that there is entanglement in the system; however, positive values of $E_W(N)$ do not guarantee separability.

## III. BRIEF SURVEY ON RARE-EARTH IONS

Rare-earth (or lanthanides) are those ions from lanthanum (La) to lutetium (Lu). Due to their similar ionic radius, scandium (Sc) and yttrium (Y) of group IIIB of the Periodic Table are considered pseudolanthanides. Lanthanides have a base configuration derived from the noble gas xenon (Xe) and have an incomplete $4f$ shell. When forming compounds, they lose $6s$ and $4f$ electrons, and the remaining unpaired $4f$ electrons are responsible for the magnetic behavior. Furthermore, the $4f$ shell is shielded by the $5s$ and $5p$ shells from the surroundings [18]; this makes possible a magnetic description in terms of localized magnetic moments with well-defined spin and orbital angular momenta.

In addition, these ions exhibit a systematic variety and complexity, which makes their magnetic properties fascinating [19]. Since we are only concerned with the magnetic properties of single rare-earth ions, the Hamiltonian of interest is

$$H_{SL} = \lambda\vec{L}\cdot\vec{S}, \tag{7}$$

where $\lambda$ is the spin-orbit coupling parameter that can be either negative or positive, for a less or more than half-filled subshell, respectively. $\vec{S}$ and $\vec{L}$ are the effective spin and orbital angular momentum obtained from Hund rules.

The spin-orbit Hamiltonian can be rewritten in terms of the total angular momentum $\vec{J} = \vec{L} + \vec{S}$, more precisely, in terms of $\vec{J}^2$, $\vec{L}^2$, and $\vec{S}^2$, and the energy eigenvalues read as

$$E_j = \frac{\lambda}{2}[j(j+1) - l(l+1) - s(s+1)], \tag{8}$$

where $j$ goes from $j = |l-s|$ to $j = l+s$. However, the $j$ multiplet corresponding to the ground state depends on the signal of $\lambda$: $j = |l-s|$ is the ground state for light rare-earth and $j = l+s$ is the ground state for heavy rare-earth. Note the energy difference between eigenstates is proportional to the spin-orbit coupling parameter $\lambda$ and, for rare-earth, the magnitude of this coupling reaches thousands of kelvins. These characteristics make rare-earth ions excellent candidates for applications in quantum-information processes at room temperature. See Table I for a further understanding and review.

## IV. MAGNETIC SUSCEPTIBILITY AS ENTANGLEMENT WITNESS: SPIN-ORBIT CASE

In Sec. II we presented an observable obtained by Wieśniak *et al.* capable of identifying entanglement between spins in a solid. However, this quantity is only valid for systems consisting of $N$ spins, all of them with the same value of an arbitrary spin length $s$ in a lattice. Another interesting work leading with entanglement and even more general quantum correlations between spins of magnitude 1, with linear and quadratic coupling between them, can be found in Ref. [20].

In this section, our purpose is to introduce a quantity to identify entanglement between angular momenta of different natures inside the same ion. In particular, we are interested in entanglement between spin and orbital angular momenta.

We start from the principle that the Hamiltonian $H_0$ of the system of interest can be described by a spin-orbit coupling between the total spin $\vec{S}$ and total orbital angular momentum $\vec{L}$, given by the Hund rules. In order to explore the magnetic properties, we apply a weak magnetic field $\vec{B}$ and the complete Hamiltonian reads as

$$H = \lambda \vec{L} \cdot \vec{S} + \mu_B(2\vec{S} + \vec{L}) \cdot \vec{B}. \tag{9}$$

Note that, due to the different $g$ factors for the spin and orbital momenta, the above Hamiltonian does not commute with the total magnetic moment, and the magnetic susceptibility is no longer related to the variance of the magnetization as in Eq. (1). In this case, the correct approach is to differentiate directly the magnetization of the system. Let us choose, for simplicity, the magnetic field along the $z$ axis. The magnetic susceptibility can be written as

$$\chi_z = \lim_{B\to 0} \frac{\partial \langle \mu_z \rangle}{\partial B_z} = \lim_{B\to 0} \frac{\partial}{\partial B_z} \left( \frac{\mathrm{Tr}(e^{-\beta H}\mu_z)}{\mathrm{Tr}(e^{-\beta H})} \right), \tag{10}$$

where $\mu_z$ is the magnetic momentum along the $z$ direction and $\mu_0 = 1$. Considering the restriction $[H,\mu_z] \neq 0$, a careful differentiation yields the relation [21]

$$\begin{aligned} \frac{\partial \langle \mu_z \rangle}{\partial B_z} &= -\int_0^\beta \left\langle e^{uH} \frac{\partial H}{\partial B_z} e^{-uH} \mu_z \right\rangle du + \beta \left\langle \frac{\partial H}{\partial B_z} \right\rangle \langle \mu_z \rangle \\ &= \int_0^\beta \langle e^{uH} \mu_z e^{-uH} \mu_z \rangle du - \beta \langle \mu_z \rangle^2. \end{aligned} \tag{11}$$

After taking the limit $B \to 0$, it is convenient to rewrite the above expression using $\mu_z = -\mu_B (J_z + S_z)$ with $\vec{J} = \vec{L} + \vec{S}$ and $[H_{SL}, J_z] = 0$, where $H_{SL}$ is the spin-orbit Hamiltonian. Thus,

$$\begin{aligned} \langle e^{uH_{SL}} \mu_z e^{-uH_{SL}} \mu_z \rangle = -\mu_B \big( \langle J_z^2 \rangle + \langle J_z S_z \rangle + \langle S_z J_z \rangle \\ + \langle e^{uH_{SL}} S_z e^{-uH_{SL}} S_z \rangle \big). \end{aligned} \tag{12}$$

After recovering the correlations in terms of $S_z$ and $L_z$, the magnetic susceptibility when the field is aligned along the $z$ axis reads

$$\begin{aligned} \chi_z = \beta\mu_B^2 \Bigg( \langle L_z^2 \rangle - \langle L_z \rangle^2 + 3 \langle S_z^2 \rangle - 4 \langle S_z \rangle^2 + 4 \langle L_z S_z \rangle \\ - 4 \langle L_z \rangle \langle S_z \rangle + \frac{1}{\beta} \int_0^\beta du \langle e^{uH_{LS}} S_z e^{-uH_{LS}} S_z \rangle \Bigg). \end{aligned} \tag{13}$$

From the above result it is possible to construct an entanglement witness in the same way as in Ref. [16]. First, let us assume that our system is prepared in a general product state, related to the $S$-$L$ bipartition. With this assumption, the terms $\langle L_z S_z \rangle$ and $\langle L_z \rangle \langle S_z \rangle$ are identical and cancel their contributions. In a second step, the derivation of the magnetic susceptibility with the magnetic field along either the $y$ axis or the $x$ axis provides expressions completely equivalent to Eq. (13) and the sum $\bar{\chi} = \chi_x + \chi_y + \chi_z$ of the susceptibilities along the three perpendicular axes, for a general product state, becomes

$$\begin{aligned} \bar{\chi} = \beta\mu_B^2 \sum_{v=x,y,z} \Bigg( \langle L_v^2 \rangle - \langle L_v \rangle^2 + 3 \langle S_v^2 \rangle - 4 \langle S_v \rangle^2 \\ + \frac{1}{\beta} \int_0^\beta du \langle e^{uH_{LS}} S_v e^{-uH_{LS}} S_v \rangle \Bigg). \end{aligned} \tag{14}$$

For an arbitrary state of a spin $S$, the relations $\langle \vec{S}^2 \rangle = s(s+1)$ and $\langle \vec{S} \rangle^2 \leqslant s^2$ are satisfied; we use the same for a state of an orbital angular momentum $l$, i.e., $\langle \vec{L}^2 \rangle = l(l+1)$ and $\langle \vec{L} \rangle^2 \leqslant l^2$. Using these relations in Eq. (14) provides the following inequality for $\bar{\chi}$:

$$\bar{\chi} \geqslant \beta\mu_B^2 [l + s(3-s) + I(l,s)], \tag{15}$$

where the integral

$$I(l,s) = \frac{1}{\beta} \sum_{v=x,y,z} \int_0^\beta du \langle e^{uH_{LS}} S_v e^{-uH_{LS}} S_v \rangle \tag{16}$$

is positive for all values of $l$ and $s$, as can be seen directly from the expression

$$\langle e^{uH_{LS}} S_v e^{-uH_{LS}} S_v \rangle = \sum_{ij} e^{u(\varepsilon_i - \varepsilon_j) - \beta\varepsilon_i} \left| \langle i \left| S_v \right| j \rangle \right|^2. \tag{17}$$

The inequality (15) is also valid for a general density operator of a separable state since it is constructed as a convex sum of product states; i.e.,

$$\begin{aligned} \bar{\chi} &= \beta\mu_B^2 [4\Delta^2(\vec{S})_\rho + \Delta^2(\vec{L})_\rho - \langle \vec{S}^2 \rangle_\rho + I(l,s)_\rho] \\ &= \beta\mu_B^2 \sum_n w_n [4\Delta^2(\vec{S})_n + \Delta^2(\vec{L})_n - \langle \vec{S}^2 \rangle_n + I(l,s)_n] \\ &\geqslant \beta\mu_B^2 \left[ l + s(3-s) + I(l,s) \right], \end{aligned} \tag{18}$$

where the index $n$ identifies the $n$th subensemble in the mixture, and $\Delta^2(\vec{J})_\rho = \langle \vec{J}^2 \rangle_\rho - \langle \vec{J} \rangle_\rho^2$ is taken in a state $\rho$. Since, for our Hamiltonian of interest, the contribution to $\bar{\chi}$ is the same for each orthogonal direction, the entanglement witness can be explicitly written as

$$E_W = \frac{3\chi_z}{\beta\mu_B^2 \left[ l + s(3-s) + I(l,s) \right]} - 1 \geqslant 0. \tag{19}$$

Therefore, if inequality (19) is not satisfied the system contains entanglement between spin and orbital angular momentum.

## V. CONNECTIONS WITH EXPERIMENTAL RESULTS

Based on the results obtained for spin dimers [22], our candidates for spin-orbit entanglement are the light rare-earth

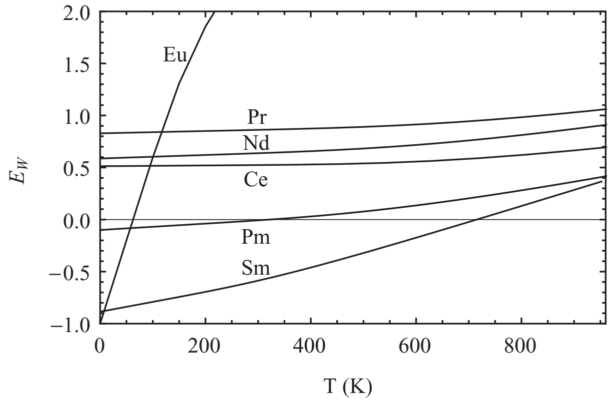


FIG. 1. Entanglement witness for light rare-earth as a function of temperature.

ions due to the antiparallel spin-orbit coupling. However, when the coupling is among angular momenta of different sizes, the explanation is not obvious as in the $\vec{S}$-$\vec{S}$ coupling. In this section we limit our results to light rare-earth that actually present spin-orbit entanglement.

In order to construct the entanglement witness, the magnetic susceptibility must be calculated. Indeed, it was done using the Van Vleck theory [23] and the values of the coupling parameter $\lambda$. These last were obtained from the energy difference $\Delta$ between the first excited state and the ground state (see Table I); $\Delta = \lambda(j+1)$ for light rare-earth and $\Delta = -\lambda j$ for heavy rare-earth [24]. Figure 1 presents the entanglement witness for light rare-earth. The witness is sensitive to the spin-orbit entanglement of Eu, Pm, and Sm. The temperature $T_E$ below which it is possible to ensure entanglement (named the entanglement temperature) is $T_E(\mathrm{Eu}) = 62$ K, $T_E(\mathrm{Pm}) = 322$ K, and $T_E(\mathrm{Sm}) = 715$ K. This result confirms, by a simple method, that at least promethium and samarium can maintain the spin-orbit entanglement up to room temperature, or even higher.

In Fig. 2 we compare the entanglement witness constructed using the magnetic susceptibility from Van Vleck theory and from experimental data of a $EuBO_3$ sample [25]. The good agreement reflects that in this case the energy spectrum is just weakly affected by the crystalline field and the Van Vleck theory is a very good approximation. In this case, the maximum temperature below which there is spin-orbit entanglement is 59 K, and the difference in comparison to Fig. 1 comes from the $\lambda_{\mathrm{Eu}} = 471$ K value used in Ref. [25]. When the crystalline field is important, the witness must be modified, a study that will be published elsewhere. Since the ground state of europium is a maximally entangled state (the only pure ground state among rare-earth), a higher entanglement temperature was expected. However, our witness seems not to be sensitive enough to the thermal spin-orbit entanglement present in europium. Probably the reason for this result is that the ground state of europium is a nonmagnetic state, since the $j$ value is null and our witness depends on a magnetic measurement.

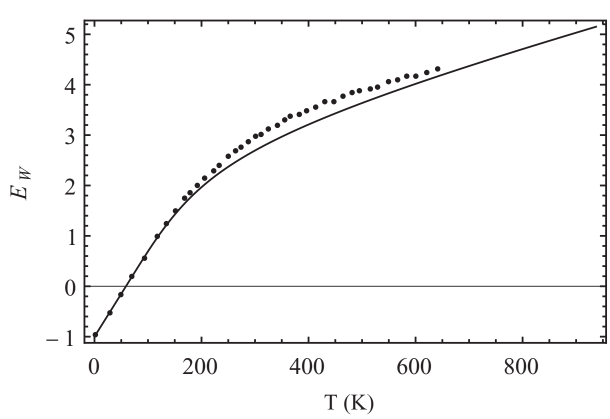


FIG. 2. Entanglement witness for the $EuBO_3$ compound as a function of temperature. The points were obtained using the experimental magnetic susceptibility from Ref. [25].

## VI. CONCLUSIONS

In this work we show that rare-earth ions can be good candidates for applications in quantum-information processes at high temperature, since some of them present spin-orbit entanglement temperatures higher than room temperature. In order to determine the entanglement, we introduce a witness, based on the magnetic susceptibility, capable of resolving entanglement between spin and orbital angular momenta. The procedure presented for deriving the entanglement witness is applicable to Hamiltonians that do not commute with the total magnetic moment. This feature makes it possible to deal with systems formed by interacting angular momenta of different size and even different natures.

Our results show that at least two rare-earth ions, promethium and samarium, maintain the spin-orbit entanglement up to temperatures close to or even higher than room temperature, which makes them excellent options for implementation of quantum processes at high temperatures. The high entanglement temperatures are due to the high values of the spin-orbit coupling parameter $\lambda$, which guarantee the system will remain in the ground state for a wide interval of temperatures.

The applicability of the witness was tested using experiment data, obtained from the literature, of a $EuBO_3$ sample. The good agreement with theoretical results justifies the assumptions made in the calculation of the magnetic susceptibility.

We know spin-orbit entanglement actually exists in Ce, Pr, and Nd as we reported in Ref. [7] using an energy-based witness. In spite of the witness (19) not showing spin-orbit entanglement for these ions, we are convinced it is valuable due to the simple and direct process involved in the experimental determination of the magnetic susceptibility.

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