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It is investigated how a transverse magnetic field affects the quantum-mechanical character of $LiHoF_4$, a system generally considered as a textbook example for an Ising-like quantum-phase transition. In small magnetic fields, the low-temperature behavior of the ions is Ising-like, involving the nearly degenerate low-lying $J_z = \pm 8$ doublet. However, as the transverse field increases, there is a substantial admixture of states having $|J_z| < 8$. Near the quantum-phase-transition field, the system is distinctively non-Ising like, and all J_z eigenstates yield ground-state contributions of comparable magnitude. A classical analog to this mechanism is the micromagnetic single point in magnets with uniaxial anisotropy. Since Ho^{3+} has $J = 8$, the ion's behavior is reminiscent of the classical limit ($J = \infty$), but quantum corrections remain clearly visible. [http://dx.doi.org/10.1063/1.4942950]

I. INTRODUCTION

The Ising quantum chain in a transverse magnetic field is a conceptually simple model with a nontrivial quantum phase transition (QPT). In contrast to Curie transitions, such quantum transitions occur at zero temperature and involve exotic quantum states rather than critical fluctuations of thermodynamic origin.[1,2] The quasi-one-dimensional tetragonal compound $LiHoF_4$ is generally regarded an experimental textbook example of an Ising-like QPT.[3–6] In the absence of a transverse magnetic field and at low temperatures (including the Curie temperature of about 1.53 K), the ground state of the Ho^{3+} ions is a well-defined and nearly degenerate crystal-field doublet ($J_z = \pm 8$), and this doublet can be mapped onto an Ising spin. However, for two reasons, the Ising character of the magnetism of $LiHoF_4$ merits a re-examination.

First, in a strict sense, there are no quantum-mechanical Ising models. By definition, Ising models have two quantum states $S_z = \pm 1/2$ (or $J_z = \pm 1/2$) per site,[7] in contrast to the $S = 1/2$ Heisenberg model, where a noninteracting spin has two eigenvalues $\pm 1/2$ but the spin can point in any direction on the Bloch sphere. The Ising model has a quasiclassical visualization as a Heisenberg spin with strong single-ion easy-axis anisotropy, but quantum-mechanically, this single-ion anisotropy is zero, because S-state ions and are therefore isotropic.[8] In fact, the Ising models usually considered in quantum-phase transitions are Heisenberg models with anisotropic exchange, which is, strictly speaking, a very different class of models.[9]

Second, Ho^{3+} is a nearly classical ion, because the relative effect of quantum fluctuations decreases approximately as $1/2J$ with increasing J. Quantum fluctuations are most pronounced for $J = 1/2$, but the Ho^{3+} ion in $LiHoF_4$ has $J = 8$ ($1/2J = 0.0625$) and should therefore behave nearly classically ($1/2J = 0$). The question therefore arises how the quasiclassical character of Ho^{3+} manifests itself in magnetic measurements.

In this paper, we scrutinize quantum-mechanical behavior of the Ho^{3+} ions. Our focus is on the atomic-scale physics of $LiHoF_4$ — no specific consideration will be given to the interatomic-coupling aspect of the material, which is crucial for the understanding of fluctuations near the phase transition but only loosely related to the atom's internal spin structure. We elaborate that the QPT

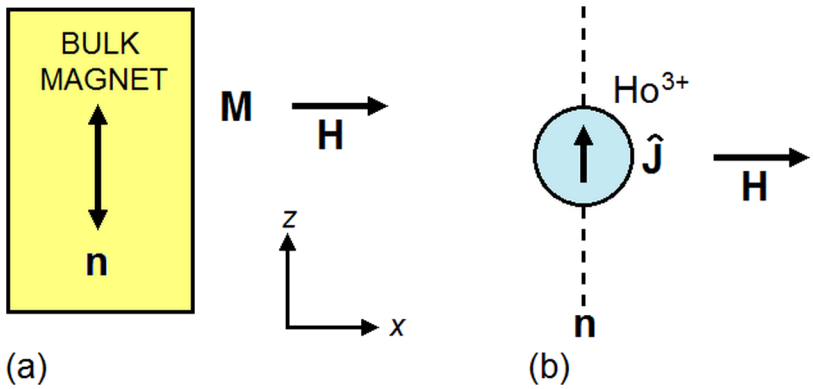

FIG. 1. Uniaxial anisotropy (**n**) and transverse magnetic field: (a) classical magnets and (b) quantum-mechanical case of a Ho^{3+} ion ($J = 8$).

singularity in a transverse field corresponds to the classical single point well-known in micromagnetism,[8] calculate the quantum corrections near the single point, and show that the behavior near the single point is non-Ising-like.

II. SINGLE-POINT ANALYSIS

Figure 1 compares the classical and quantum-mechanical limits of anisotropic magnets in a transverse magnetic field. In the classical limit, the Hamiltonian is of the Stoner-Wohlfarth type

$$\mathcal{H} = -\frac{1}{3}K_1V(3\cos^2\theta - 1) - \mu_o M_s H_x V \sin\theta - \mu_o M_s H_z V \cos\theta \tag{1}$$

where θ is the magnetization angle, defined by $M_z = M_s \cos\theta$ and $M_x = M_s \sin\theta$. The quantities K_1, M_s, **H**, and V have their usual meanings, namely lowest-order uniaxial anisotropy constant, saturation magnetization, applied magnetic field, and magnet volume, respectively.

Minimizing Eq. (1) for $\mathbf{H} = H_x\,\mathbf{e}_x$ and monitoring M_x yields the solid curve of Fig. 2. The curve exhibits a singularity at the anisotropy field $H_A = 2K_1/\mu_o M_s$. This singularity is exploited in a well-established experimental method known as single-point detection (SPD),[10] which traces the second derivative d^2M_z/dH^2. Ideally, this derivative is a delta peak, but in practice, it is a simple maximum near H_A, because sample and grain misalignments smooth the singularity (dashed line in Fig. 2). This effect was first noticed the original paper by Stoner and Wohlfarth.[11] The SPD method can be used to experimentally determine magnetic anisotropies, for example by using a pulse field.

From the classical relations $M_x(H \le H_A) = M_s\,H_x/H_A$ and $M_s^2 = M_x^2 + M_z^2$ it follows that

$$M_z = \sqrt{M_s^2 - M_x^2} \tag{2}$$

Figure 2 shows the field dependences of M_x and M_z. The blue curve (M_z) is a classical analog of the quantum-phase transition, whereas the red curve (M_x) is the same as the solid curve in Fig. 2.

III. QUANTUM-MECHANICAL LIMIT

In suitable dimensionless units, the Hamiltonian of the Ho^{3+} ion subjected to a second-order uniaxial crystal field ($A_2^0 \sim B_{20}$) and an external magnetic field (H_x) is

$$\mathcal{H} = -(3\,\hat{J}_z^{\,2} - J(J+1)) - h\hat{J}_x \tag{3}$$

where $\hat{\boldsymbol{J}}$ is the angular-momentum operator of the Ho^{3+} ion ($J = 8$) and $h \sim H_x/A_2^0$. It is convenient to use a basis where $\hat{J}_z$ is diagonal, with the eigenvalues $J_z = (-8,\ldots,7,8)$. In this basis, $\hat{J}_x$ is

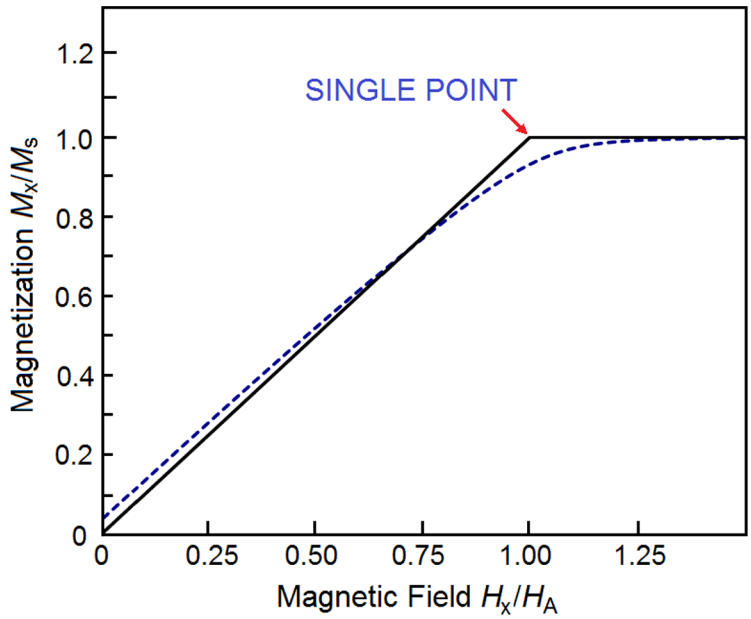

FIG. 2. Approach to saturation and single-point detection. A true singularity (solid curve) exists for perfectly aligned magnets only ($\mathbf{n} \perp \mathbf{H}$). Any oblique angles, such as 2° in the case of the dashed line, smooth the singularity.

tridiagonal and symmetric with the matrix elements $\hat{J}_x(j,j) = 0$ and

$$\hat{J}_x(j,j+1) = \frac{1}{2}\sqrt{72-(8-j)(7-j)} \tag{4}$$

Note, in particular, that the matrix element connecting the states with $J_z = \pm 8$ is zero. The eigenvalues and eigenfunctions of Eq. (3) are readily obtained by explicit matrix diagonalization.

In zero field, Eq. (3) yields the double-degenerate ground state, $J_z = \pm 8$, of energy $E_g = -120$. The energy-level splitting between the ground-state and first excited doublets is 45 energy units.

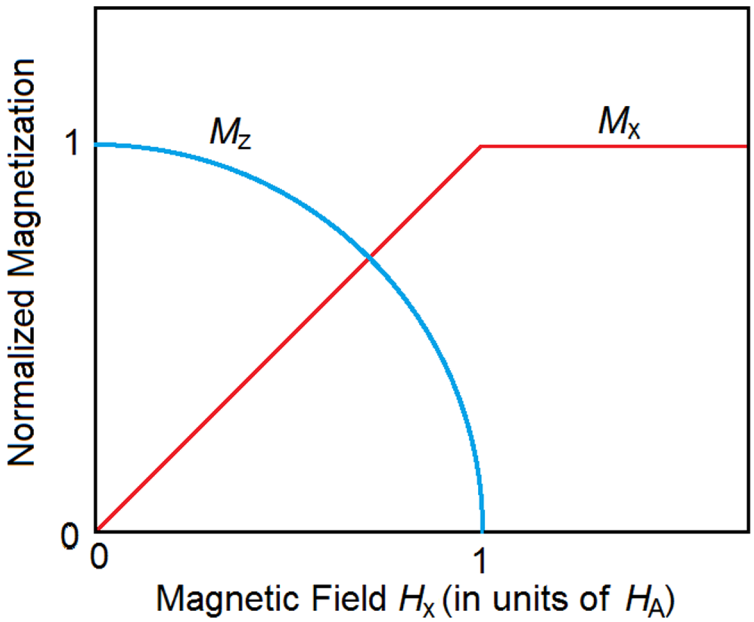

FIG. 3. Relation between single point and vanishing of the magnetization in the classical limit ($J > \infty$). The field where the magnetization M_z vanishes is equal to the single-point field (anisotropy field).

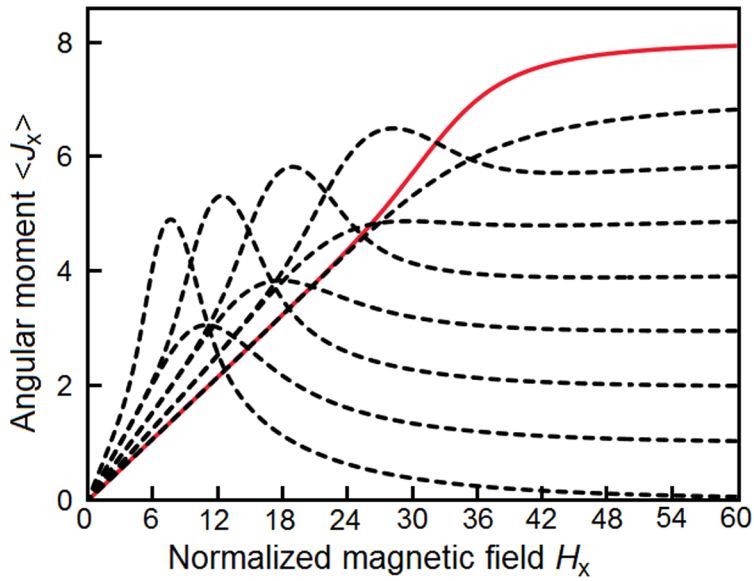

FIG. 4. Energy levels calculated from the Hamiltonian of Eq. (1). The solid red curve refers to the single-ion ground state, whereas the dashed lines are excited one-ion levels.

This splitting is much larger than the interaction strength between the Ho^{3+} ions, which is epitomized by the Curie temperature, 1.53 K for $LiHoF_4$.[3] Excited levels can therefore be ignored at all temperatures of interest, and the system behaves like an Ising spin with two spin orientations ($s_z = \pm 1$).[2–4] However, this argumentation applies to small fields only, not to the transverse fields that cause the magnetization M_z to vanish at the zero-temperature (or low-temperature) quantum-phase transition.

Figure 4 shows the expectation values <J_x> as a function of the transverse field H_x. The red curve refers to the ground state, whereas the dashed curves correspond to excited states. In the limit of very high transverse fields, the levels are equally spaced Zeeman levels. Comparison of the solid

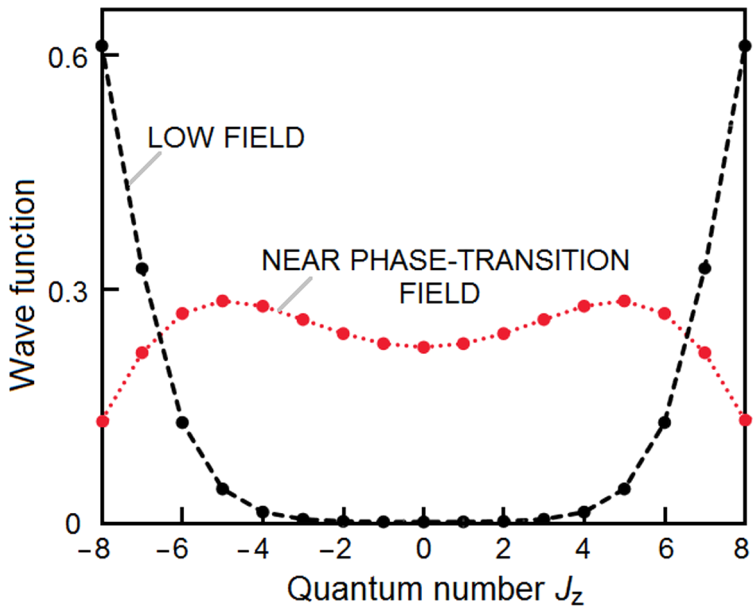

FIG. 5. Quantum mechanical approach to saturation in the field direction. In terms of Fig. 4, the applied fields are $h = 12$ (dashed black curve) and $h = 36$ (dotted red curve).

red lines in Figs. 3 and 4 shows that the approach to saturation is nearly classical, although the effect of quantum fluctuations is clearly visible in Fig. 4.

Figure 5 compares the ground-state eigenfunctions in low transverse fields ($h = 12$) and near the single point ($h = 36$). In low fields, the system remains Ising like, dominated by states with $J_z = \pm J$ and having a relatively small admixture of states with $|J_z| < 8$. However, near the single point, the ground state is no longer Ising like but represents a superposition of all J_z states.

IV. DISCUSSION AND CONCLUSIONS

Our analysis ignores the dipolar and exchange interactions between the Ho ions. These interactions are important for the understanding of the quantum-phase transition, but they do not affect the present findings, because they do not undo the mixing of the quantum states near the phase transition. In fact, the interactions do not interfere in lowest order, because ensembles of ions described by Eq. (3) rotate coherently, that is, little or no penalty is imposed through changing angles between neighboring Ho moments. Furthermore, the interactions are weaker than the level splitting caused by the anisotropy.[4] Similarities between classical and quantum-mechanical predictions, as epitomized by the red curves in Figs. 3 and 4, respectively, indicate the need to study quantum-phase transitions on a materials-specific basis.

In summary, we have investigated the effect of a transverse magnetic field on $LiHoF_4$. In the classical limit, the quantum-phase transition assumes the character of a micromagnetic single point. Since the Ho^{3+} ions have $J = 8$, their behavior is reminiscent of classical spins, but quantum effects remains clearly visible in the magnetization curves. The transverse-field Ising model captures some nontrivial features of quantum-phase transitions, but it does not provide an adequate description of $LiHoF_4$, which has been considered an archetypical Ising system. In small transverse fields, the system is Ising-like, but the Ising character wanes as the transverse magnetic field increases. Our analysis indicates that a comprehensive and materials-specific analysis of quantum phase transitions in systems with $J > 1/2$ remains a challenge to future research.

ACKNOWLEDGEMENT

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