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Turbulent fronts of quantum detonation in molecular magnets

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Dipolar-controlled quantum deflagration going over into quantum detonation in the elongated Mn_{12}Ac molecular magnet in a strong transverse field has been considered within the full three-dimensional model. It is shown that within the dipolar window around tunneling resonances the deflagration front is nonflat. With increasing bias, dipolar instability makes the front turbulent, while its speed reaches sonic values, which is a signature of detonation.

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Magnetic burning or deflagration, similar to the usual chemical burning, has been experimentally observed on elongated crystals of molecular magnet (MM) Mn_{12}Ac in Refs. 1,2. Deflagration is controlled by the temperature rising as the result of the relaxation from the metastable to stable state of this bistable system (burning), accompanied by the heat conduction toward the cold unburned region. This leads to the formation of a flat burning front moving at a constant speed controlled by the energy release set by the bias magnetic field B_z applied along the easy axis of magnetic molecules. This discovery triggered theoretical³ and further experimental^{4,5} research. In these experiments no transverse field was applied and the front speed was as low as 1–15 m/s. Fast moving burning fronts in Mn_{12}Ac initiated by a fast magnetic field sweep have been observed in Ref. 6. In this region, deflagration can go over into detonation with a speed comparable to the speed of sound.⁷ Crossover from relaxation to deflagration in Mn_{12}Ac in a strong transverse field has been experimentally studied in Ref. 8.

Since MMs are famous exponents of the resonant spin tunneling^{9,10} (see Ref. 11 for a review), manifestations of the latter were expected in magnetic deflagration. Indeed, maxima of the front speed at tunneling resonances were seen in Refs. 2,4. The simplest way to explain these maxima is to use, within the standard deflagration theory, the Arrhenius relaxation rate $\Gamma(B_z, T)$ having maxima at B_z corresponding to tunneling resonances.^{2,3}

It was further suggested that in the case of a strong tunneling directly out of the metastable ground state, caused by a strong transverse magnetic field, fronts of nonthermal quantum or “cold” deflagration are possible.^{12,13} Here, instead of the temperature, tunneling is controlled by the dipolar field of the crystal that can block and unblock tunneling by setting magnetic molecules on or off resonance. Within the simplified one-dimensional (1D) approximation it was shown that the magnetization in the moving front is adjusting self-consistently, even in the presence of disorder,¹² so that the dipolar field unblocks tunneling within the front core. The theory of quantum deflagration has been generalized^{14,15} to include both quantum and thermal effects via the relaxation rate $\Gamma(B_z, T)$ numerically calculated from the density-matrix equation.¹⁶ The main finding was a strong quantum acceleration of the front within the dipolar window around the resonance values of the external field $B_z = B_k$:

$$B_k - B_z^{(k_D)} \leq B_z \leq B_k + B_z^{(D)}, \quad (1)$$

where $B_z^{(D)} = 52.6$ mT is the dipolar field produced by the fully magnetized long crystal and $B_z^{(k_D)} = 72.9$ mT.¹⁵ The front speed was supersonic and apparently diverging towards the right end of the dipolar window, see Fig. 15 of Ref. 15 and discussion therein. Indeed, as the dipole-dipole interaction (DDI) is instantaneous, there is no limitation on the front speed.

While fast quantum deflagration in MM is awaiting experimental confirmation, a theoretical problem remains. The 1D approximation assumes that the magnetization and dipolar field in the front depend only on the coordinate z along the sample (which coincides with the MM easy axis). This is a crude approximation, and there should be a dependence on x and y that renders the front nonflat. Moreover, dipolar-controlled front should be unstable at a small scale because portions of the front that are ahead of the neighbors create dipolar field on the latter that hampers their motion, while the lagging neighbors create a field that accelerates the leader (see Fig. 1). This instability reflects the fact that a long MM crystal tends to split up into domains parallel rather than perpendicular to the z axis.¹⁷ As the result, the front can become turbulent with a chaotic dipolar bias rendering many magnetic molecules inside the front core off resonance. The question is whether the front speed drops to that of the regular slow burning^{1,18} or remains fast. In the latter case one has to speak of quantum detonation rather than of quantum deflagration.

The purpose of this paper is to present a full 3D theory of fronts of tunneling in molecular magnets and clarify the role of the dipolar instability.

The system of equations describing deflagration with quantum effects in molecular magnets¹⁴ includes the rate equation describing relaxation of the metastable population n ($0 \leq n \leq 1$)

$$\dot{n}(t, \mathbf{r}) = -\Gamma[B_{\text{tot},z}(\mathbf{r}), T(\mathbf{r})][n(t, \mathbf{r}) - n^{(\text{eq})}]. \quad (2)$$

Here the relaxation rate Γ depends on the total bias field

$$B_{\text{tot},z}(\mathbf{r}) = B_z + B_z^{(D)}(\mathbf{r}) \equiv B_z + (Sg\mu_B/v_0) D_{zz}(\mathbf{r}), \quad (3)$$

(external plus dipolar field) and the temperature at a given point, while $n^{(\text{eq})}(T)$ is the equilibrium metastable population (set to zero below). Here $v_0 = a^2c$ is the unit-cell volume, a and c are lattice spacings, and $D_{zz}(\mathbf{r})$ is the reduced dipolar field. Another equation is the heat conduction equation that can be conveniently written in terms of the thermal energy $\mathcal{E}$ per magnetic molecule

$$\dot{\mathcal{E}}(t, \mathbf{r}) = \nabla \cdot \kappa \nabla \mathcal{E}(t, \mathbf{r}) - \Delta E \dot{n}(t, \mathbf{r}). \quad (4)$$

Dipolar instability of a flat front of spin tunneling

FIG. 1. (Color online) Dipolar instability of a smooth front of tunneling. The leading parts of the front (center) are helped by the lagging neighbors while hampering the motion of the latter.

Here $\Delta E \cong 2Sg\mu_B B_z$ in the source term of this equation is the released energy per molecule, κ is thermal diffusivity (a crude estimate $\kappa \cong 10^{-5} \text{ m}^2/\text{s}$ ^{1,5}). $\mathcal{E}$ and T are related via the measured¹⁹ heat capacity $C = d\mathcal{E}/dT$.

While the Eqs. (2) and (4) are the same as the standard deflagration equations,³ the resonance form of Γ and its crucial dependence on the dipolar field $B_z^{(D)}(\mathbf{r})$ that is defined by $n(t, \mathbf{r})$ makes a big difference. For the realistic model of Mn_{12}Ac with the uniaxial part of the Hamiltonian $-DS_z^2 - AS_z^4$, tunneling resonances are achieved at $B_{\text{tot},z} = B_{km}$, where

$$g\mu_B B_{km} = k[D + (m^2 + (m+k)^2)A], \quad (5)$$

$k = 0, 1, 2, \dots$, and $m = -S, -S+1, \dots$ label the metastable spin states. The small fourth-order anisotropy A splits k resonances into m multiplets. With Mn_{12}Ac parameters from Ref. 20 and using the density-matrix-equation method of Ref. 16, for the transverse field $B_{\perp} = 3.5 \text{ T}$ one obtains Γ around the first resonance, $k = 1$, shown in Fig. 2. At such strong $B_{\perp}$ only the resonances with $m = -10$ (ground-state resonance) and $m = -9$ are seen, while all the others are broadened away.

The most challenging part of the work is numerical calculation of the dipolar field produced by the sample. $D_{zz}(\mathbf{r})$ in Eq. (3) is defined by the lattice sum $D_{zz}(\mathbf{r}_i) = \sum_j \phi(\mathbf{r}_j - \mathbf{r}_i)\sigma_z(\mathbf{r}_j)$, where

$$\phi(\mathbf{r}) \equiv v_0[3(\mathbf{e}_z \cdot \mathbf{n})^2 - 1]/r^3, \quad \mathbf{n} \equiv \mathbf{r}/r. \quad (6)$$

FIG. 2. (Color online) Relaxation rate of Mn_{12}Ac vs temperature and longitudinal magnetic field in the transverse field $B_{\perp} = 3.5 \text{ T}$.¹⁵ One can see the ground-state resonance at $B_z = 0.522 \text{ T}$ and the first-excited-state resonance at $B_z = 0.490 \text{ T}$ for $k = 1$ multiplet.

To calculate this sum, one can introduce a small sphere of radius r_0 around $\mathbf{r}_i$ satisfying $v_0^{1/3} \ll r_0 \ll L$, where L is the (macroscopic) size of the sample. Assuming that $\sigma_z(\mathbf{r})$ does not change at the scale r_0 , one obtains

$$D_{zz}(\mathbf{r}) = \frac{v}{v_0} \int_{|\mathbf{r}' - \mathbf{r}| > r_0} d\mathbf{r}' \phi(\mathbf{r}' - \mathbf{r}) \sigma_z(\mathbf{r}') + \sigma_z(\mathbf{r}) \bar{D}_{zz}^{(\text{sph})}, \quad (7)$$

where $v = 2$ is the number of magnetic molecules per unit cell v_0 for the body-centered tetragonal Mn_{12}Ac and the lattice-dependent $\bar{D}_{zz}^{(\text{sph})} = 2.155$ comes from summation over the small sphere. The restriction on the integration can be removed by subtracting $\sigma_z(\mathbf{r})$ from the integrand and adding the counterterm. Finally one obtains

$$D_{zz}(\mathbf{r}) = \frac{v}{v_0} \int d\mathbf{r}' \phi(\mathbf{r}' - \mathbf{r}) [\sigma_z(\mathbf{r}') - \sigma_z(\mathbf{r})] + \sigma_z(\mathbf{r}) [v \bar{D}_{zz}(\mathbf{r}) - k_D], \quad (8)$$

where $k_D \equiv 8\pi v/3 - \bar{D}_{zz}^{(\text{sph})} = 14.6$. $\bar{D}_{zz}(\mathbf{r})$ is the reduced magnetostatic field given by an integral over the sample surface. For a rectangular sample with dimensions $2L_x \times 2L_y \times 2L_z$ and $-L_x \leq x \leq L_x$ etc. one has²¹

$$\bar{D}_{zz}(\mathbf{r}) = \sum_{\eta_x, \eta_y, \eta_z = \pm 1} \arctan \frac{(L_x + \eta_x x)^{-1} (L_y + \eta_y y) (L_z + \eta_z z)}{\sqrt{(L_x + \eta_x x)^2 + (L_y + \eta_y y)^2 + (L_z + \eta_z z)^2}} \quad (9)$$

+ ($x \Rightarrow y$), in total 16 different arctan terms. Inside a uniformly magnetized long Mn_{12}Ac crystal Eq. (8) yields $D_{zz} = 10.53$ that in real units corresponds to $B_z^{(D)} = 52.6 \text{ mT}$. The same value of the dipolar field follows from the experiment.²²

For numerical solution, box-shape samples with dimensions $L_x \times L_y \times L_z$ and $0 \leq x \leq L_x$ etc. are considered, and the problem is discretized with a rectangular grid. This makes the integral in Eq. (8) a sum that can be calculated using the fast Fourier transform (FFT). In this work, numerical solution was implemented in WOLFRAM MATHEMATICA, whereas the dipolar sum was computed using FFT-based and very fast LISTCONVOLVE command. After discretization of the Laplace operator in Eq. (4) the system of equations for quantum deflagration becomes a system of nonlinear ordinary differential equations. It has been solved by the fixed-step compiled fourth-order Runge-Kutta method and, preferred, by somewhat more efficient fifth-order Butcher's Runge-Kutta method making six evaluations per step. Instead of $\mathcal{E}$, it is more convenient to use $\mathcal{E}/\Delta E + n$ as one of unknown functions. The grid sizes were $64 \times 64 \times 300$, $100 \times 100 \times 200$, and $100 \times 100 \times 300$. To reduce the amount of calculations, symmetry conditions within xy planes have been imposed.

Deflagration was ignited in the uniformly magnetized state $n = 1$ at the left end of the crystal, $z = 0$, by a quick temperature rise at the boundary. At higher B_z , deeper into the dipolar window, the resonance condition is fulfilled at a surface close to the left end, so that quantum deflagration begins spontaneously there. The right end of the sample was protected from spontaneous ignition by adding a semi-infinite fictitious sample with a fixed down magnetization, $n = 1$. The sample

FIG. 3. (Color online) Front speed within the 3D model for a strong transverse field ($B_{\perp} = 3.5$ T) in the vicinity of the ground-state tunneling resonance at $B_z = B_k = 0.522$ T.

was considered as thermally insulated, the initial temperature being almost zero. The front speed was determined from the time needed for the front to arrive at the right end or from the time dependence of the average metastable population. Computations were done for $L_x = L_y = 0.2$ mm⁴ and 1 mm.² One burning event required about one day computations on a workstation.

The results for the front speed in the vicinity of the first tunneling resonance ($k = 1$) are shown in Fig. 3. Beyond the dipolar window, $B_z < 0.45$ T and $B_z > 0.573$ T, there is no tunneling and the usual temperature-driven deflagration with a smooth flat front and $v \approx 50$ m/s takes place. Entering the dipolar window from the left leads to a gradual increase of the effect of tunneling. The front speed increases and reaches sonic values, diverging at the right end of the dipolar window $B_k + B_z^{(D)} = 0.522 + 0.0526 = 0.573$ T, where B_k is the ground-state tunneling resonance and $k = 1$. Although the front speed in the 3D model is lower than in the 1D model, qualitatively the results are similar (cf. Fig. 15 of Ref. 15). The front speed is higher in the thicker sample, which is not surprising as the speed scales with the sample thickness in the case of cold deflagration.^{12,13} There is no contribution of the excited-state tunneling resonance that within the 1D model leads to a front speed peak at $B_z \approx 0.42$ T in Fig. 15 of Ref. 15. The apparent reason is the dependence of the dipolar field on the transverse coordinates even in a flat front that leads to broadening of tunneling peaks.

The structure of the front of tunneling is shown in Figs. 4 and 5 and in the Supplemental Material.²³ Here n is represented as a function of x and z at $y = 0$. Color coding is blue for the cold unburned regions ($n \approx 1$) and red for hot burned regions ($n \approx 0$). Dipolar effect makes the front progressively nonflat, its central part leading as the speed is increasing, as seen in the upper image at $B_z = 0.50$ T. Also with increasing B_z the front becomes gradually nonsmooth. However, thermal burning tends to smoothen the roughness created by the dipolar instability discussed above as large temperature gradients lead to increased heat conduction and equilibration along the front. Whereas in the absence of thermal effect (cold deflagration) there is a spatially irregular unburned metastable

FIG. 4. (Color online) Profile of the metastable population n in the 3D model of fronts of tunneling in a 0.2 mm Mn_{12}Ac crystal at $B_{\perp} = 3.5$ T and $B_z = 0.5$ T (upper) and 0.56 T (lower).

population behind the front,^{12,13} here a complete burning is achieved.

Closer to the right border of the dipolar window the front becomes turbulent and looks like precipitation, as seen in the lower image in Fig. 4 for $B_z = 0.56$ T. Given its sonic speed, which cannot be provided by any thermal mechanism, one can speak of quantum detonation. The relaxation event in the figure began spontaneously on a surface near the left end where the resonance condition was satisfied. From there a regular slow-burning front is moving to the left and a quantum detonation front is going to the right.

Although in the quantum detonation front the dipolar bias is wildly changing in space so that the system does not stick to the resonance (which was the main argument in the 1D theory of fronts of tunneling^{12,13}), tunneling is still very strong to ensure sonic front speeds. This requires a closer investigation. Still, one can argue that dynamic crossing the resonance can facilitate transitions via Landau-Zener effect.

To summarize, a full three-dimensional theory of dipolar-controlled quantum deflagration with the temperature effect, going over into quantum detonation, has been proposed. The quantum front can reach sonic speeds near biased ground-state tunneling resonances and is nonflat and turbulent. To activate

FIG. 5. (Color online) Profile of the metastable population n in the 3D model of quantum deflagration in a 1 mm Mn_{12}Ac crystal at $B_{\perp} = 3.5$ T and $B_z = 0.55$ T.

quantum transitions out of the metastable ground state, a strong transverse field has to be applied. An experimental challenge is to prepare the initial state that has a limited life time.

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- ²³See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.88.064413> for videos of propagating fronts.