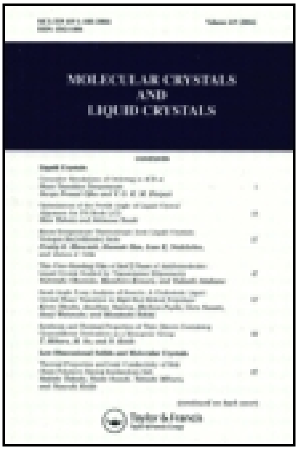

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Theoretical Studies on Quantum Tunneling of Spins in Cluster of Clusters

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In this work, we consider a cluster, which consists of small clusters with an anisotropy energy and suppose that the Hamiltonian of each cluster in the cluster is represented by the nonlinear sigma model. We apply a single spin model to the model clusters and calculate the time evolution of spin wavepacket. We demonstrate a magnetic field induced tunneling and elucidate its mechanism.

Keywords: quantum tunneling of magnetization; wavepacket dynamics; cluster of clusters

INTRODUCTION

Clusters of metal atoms or metal assembled systems have attracted a great deal of interest in recent years because of the fact that many of their properties (structural, electronic, optical, and magnetic) differ from those of the corresponding bulk. Particularly, the magnetic properties of small transition metal clusters are attractive from a technological point of view, because they can be applied to molecular memories or nano structure devices.

Quantum tunneling of magnetization has been observed in high spin state (S = 10) of manganese acetate (Mn_{12}-Ac) molecules [1,2]. It appears in steps in the hysteresis loop of oriented Mn_{12}-Ac crystals. Many experimental and theoretical efforts have been made to reveal the tunneling mechanism [3,4]. In previous papers [5,6], we have proposed a theoretical foundation on the quantum tunneling in magnetic clusters such as Mn_{12}-Ac and etc. by means of the spin coherent state path integral approach and derived a formulation of the tunneling rate.

Quantum dynamics of spin systems have been applied to investigate tunneling process between metastabel states to stable states in relation to the nonadiabatic transition and a sweep rate of applied field [7,8]. In recent work [9], we have proposed a path integral spin-centroid molecular dynamics method to investigate quantum tunneling of magnetization of S=1 system. It is, however, hardly applicable to treat quantitatively with relaxation process including effects of energy diffraction induced by interactions with thermal modes. We here focus only on a quantum tunneling of magnetization with time independent magnetic filed. In this work, we consider a model cluster, which consists of two different type clusters, as shown in Fig.1. We investigate tunneling process of magnetization of the model system by calculating real time dynamics of a single spin model.

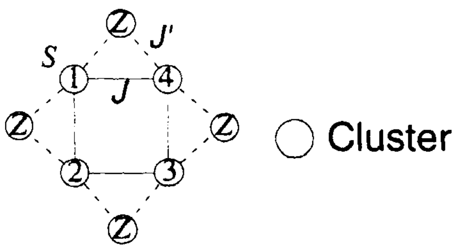


Figure 1 Model system of cluster of clusters.

THEORETICAL BACKGROUND AND CALCULATIONS

Here, we consider a cluster, which consists of clusters with the anisotropy energy, as shown in Fig.1 and suppose that the Hamiltonian of each cluster in the cluster is represented by the simple model, the nonlinear σ model:

$$H_a = -DS_{az}^2 - \mathbf{B}\cdot\mathbf{S}_a \,, \tag{1}$$

where D indicates the anisotropy energy, and $\mathbf{B}$ is the external magnetic field, assumed to be in the direction at z-axis. The total Hamiltonian of the system shown in Fig.1 may be supposed to be written as

$$H = \sum_{i=1}^{4}\left[J\mathbf{S}_a\cdot\mathbf{S}_{a+1} + J'\,\mathbf{S}_a\cdot\mathbf{S}_Z + J'\,\mathbf{S}_{a+1}\cdot\mathbf{S}_Z + H_a + H_Z\right] \tag{2}$$

Note that the anisotropy energy D is independent of the interactions between clusters in the cluster and the external magnetic field. The state of a spin at a given site is expressed with the following coherent basis:

$$|\Omega\rangle = e^{-i\phi(\hat{S}_z - S)}e^{-i\theta\hat{S}_y}|S\rangle, \tag{3}$$

where Ω is a vector on a unit sphere with spherical coordinates $(1\,,\theta\,,\phi)$, $0\le\theta\le\pi$ and $0\le\phi\le 2\pi$, and $|S\rangle$ is the eigenstate of $\hat{S}_z$ with the largest possible eigenvalue S. Using the coherent state of Eq.(2), we can reduce many-body problem to single spin model. Thus, we obtain the effective Hamiltonian of the ground state for the two cases:

$$H_{\text{eff}} = M\dot{\theta}^2 - 4(D-2J)\,S^2\left(\cos\theta - \frac{2J'S-B}{2(D-2J)S}\right)^2 + \frac{(2J'S-B)^2}{(D-2J)} - 4(DS+JS+B)S, \qquad (D-2J\neq 0) \tag{4-a}$$

$$H_{\text{eff}} = M\dot{\theta} - (2J'S-B)\,S\cos\theta - (3JS+B)S, \quad (D-2J=0) \tag{4-b}$$

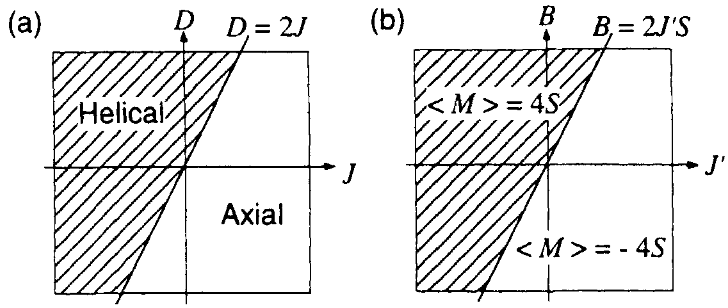


Figure 2 (a) Phase diagram of the spin structure.
(b) Phase diagram of the magnetization when D-$2J$<0.

where M denotes a mass of the collective motion of the spin rotation. From Eq.(4), it is found that when D - $2J$ is positive, the spin structure becomes axial, and when it is negative, it is helical, shown in Fig. 2-(a). Fig 2-(b) shows the phase diagram of magnetization when the spin structure is axial.

RESULT AND DISCUSSION

We evaluate real time evolution of spin wavepacket by quantizing the classical effective Hamiltonian of Eq. (4-a) when $\phi = 0$ for three extreme cases: (i) $B=0$, (ii) $B=2J'S$, (iii) $B=4J'S$, in order to investigate quantum tunneling of magnetization of axial spin structure, D-$2J$<0, at zero temperature. For an initial wavepacket of the spin coherent state, we use a Gaussian wavepacket with exponent 2.0 set at $\theta = 0$ for the cases of (ii) and (iii) or π for (i). We set $S=1.0$, $D=-1.0$, $J=1.0$, $J'=0.5$. Time dependent magnetization at several amplitude of magnetic field are depicted in Figure 3. We found that a tunneling induced by external magnetic field occurs for the case of (ii) and dose not for the cases of (i) and (iii), because the shape of the potential energy of (ii) is double minimum

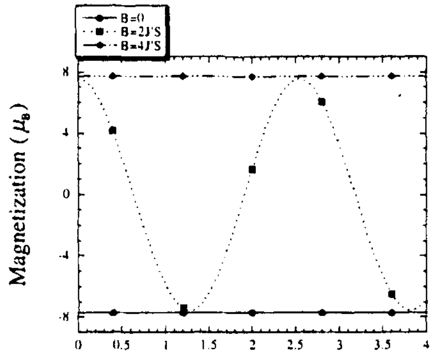
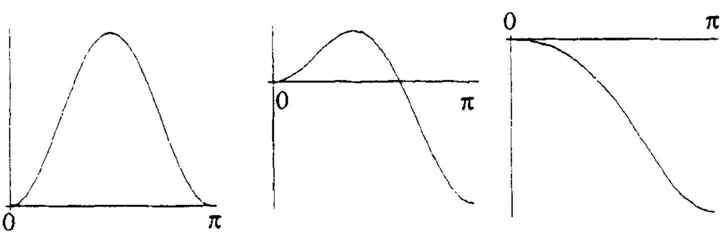


FIGURE 3 Magnetization as a function of time.

shown in Figure 4 (a). This tunneling is due to resonance of two degenerate states localized at each minimum. In the case of 4-(b), the initial wave packet is a classically metastable state and different tunnelings should be observed when the state is on resonant with other energy levels. Controlling these phenomena is discussed in future works. The more the amplitude of applied field decrease, the energy barrier becomes lower and vanishes at B=0 as shown in 4-(c). On the other hand in sufficiently large magnetic filed, say B=4J'S, the energy barrier also vanishes and the initial wave packet localized around $\theta = 0$ is a stable state. The magnetization does not switch to negative as shown in Figure 3. Thus we demonstrate that we can control the magnetic tunneling by tuning the applied magnetic field.



(a) $B=2J'S$ (b) $B=1.6J'S$ (c) $B=0$

FIGURE 4 Potential energy surface of model system.

CONCLUDING REMARKS

We first reduce a many spin problem into one corrective motion of spins by using the spin coherent state of Eq.(2). Next, we investigate magnetic tunneling process in the cluster of clusters in relation to magnitude of magnetic interaction and applied field by using wave packet dynamics with effective Hamiltonian. Particular we observed a resonant tunneling from S=4 to S=-4 states occurs when $D\text{-}2J=0$. We elucidate the difference of tunneling mechanisms by analyzing shapes of the potential energy surface. It suggests that the tunneling can be controlled by modulating the applied field and the mechanism is applicable to a nano-sized switching device.

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