Journal of Physics: Conference Series **592** (2015) 012105 doi:10.1088/1742-6596/592/1/012105

Photo-induced Insulator-ferromagnetic Metal Transition and Magnetic Solitons in the Perovskite GdSrMnO

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Abstract. We have discussed the dynamics of photo-induced magnetic solitons in the perovskite GdSrMnO by using quantized massive gauge fields, which are introduced theoretically in the path-integral method. For the first time, we propose the magnetic relaxation mechanism of photo-induced magnetic solitons by using the effective Hamiltonian with the p-magnetic solitons interaction and the Langevin equation.

1. Introduction

In the perovskite manganites, charge, spin, orbital, and lattice degrees of freedoms are strongly connected to each other, showing various kinds of the electronic phase, such as charge ordered and orbital ordered insulator, ferromagnetic metal, antiferromagnetic insulator, and so on. Tokura [1,2] has reported the photo-induced insulator-metal transition in the perovskite manganites. The photo-excitation above the charge gap in the charge-orbital ordered state can cause the hopping of the electrons or holes into the neighboring site, hence forming magnetic solitons in the regular charge-orbital ordered state. Miyano et al.[3] and Fiebig et al.[4] have reported the photo-induced transition between the antiferromagnetic insulator and the ferromagnetic metal in the manganites. Matsubara et al.[5] have investigated the ultrafast spin and charge dynamics in the course of a photo-induced phase transition from an insulator with short-range charge order and orbital order to a ferromagnetic metal in perovskite-type PrCaMnO. Ishikawa and coworkers[10-14] have studied theoretically the photo-induced phase transition in the perovskite manganites. The photo-induced dynamic magnetic effect has been studied in the II-VI-based diluted magnetic semiconductors (DMS) and III-V-based DMS, and interesting phenomena such as the photo-induced magnetic polaron have been discussed [6]. These works stimulated us to the study of the carrier-induced magnetic solitons, which is an interesting and challenging subject. Kanazawa[7-9] has discussed the insulator-metal trasnsition and large magnetoresistance effects in DMS, using the gauge-invariant Lagrangian density for the magnetic solitons. Kanazawa[15] has discussed the percolation-like insulator-metal transition, the conduction mechanism, and localization of photo-induced magnetic solitons with holes in the perovskite PrCaMnO. Recently Kanazawa[16] has proposed the percolation-like dynamics of hole-induced magnetic solitons and the colossal magnetoresistance mechanism in doped manganites from the standpoint of the random resistor network and percolation-scaling method. In addition, the long-relaxation

behavior of the spin has been discussed by using the effective Hamiltonian[16] and the frustration-limited domain[17,18]. López et al.[19] have reported the glassy behavior such as slow relaxation in the magnetization in $La_{1-x}Ca_xMnO_3$. In the series $(La_{1-x}Tb_x)_{2/3}Ca_{1/3}MnO_3$, Teresa et al.[30] have proposed the existence of two different volume states associated with the metalliclike ferromagnetic state and the semiconductorlike paramagnetic, spin-glass, or antiferromagnetic state, respectively. Small-angle neutron scattering experiments[30] reveal the existence of magnetic solitons for $x \leq 0.33$. The magnetic correlation length diverge at Tc for $x \leq 0.25$ while magnetic clusters of around 18 Å stabilized in the $x = 0.33$ compound at low temperature. Muon spin relaxation experiments confirm the absence of microscopic local magnetic order for the $x = 0.33$ compound and give evidence for the existence of static local fields randomly oriented below $\sim$ 44 K, bringing about a glassy magnetic state below the temperature. In the case of $La_{0.7-x}Y_xCa_{0.3}MnO_3$ manganite[20], the slow relaxation is interpreted as due to ferromagnetic clusters, which grow with decreasing temperature down to a temperature at which they freeze due to several intercluster frustration. This might correspond to the cluster glass. Recently Sasaki and Kanazawa[21] have proposed the mean field theory of the cluster glass mechanism and introduce theoretically the spin-glass temperature T_g of doped manganite $(Sm_{1-x}Cd_y)_{0.55}Sr_{0.45}MnO_3$. The possibility of a new kind of glass and slow spin-relaxation in manganites is a challenging and quite interesting issue that deserves further experimental and theoretical work. In this study, for the first time we shall propose the magnetic relaxation mechanism of photo-induced magnetic solitons in the perovskite GdSrMnO, extending the previous formula[15].

2. A model system and dynamics of magnetic solitons

Several optical data[31,32] present evidence of small magnetic polarons (magnetic solitons) above the ferromagnetic ordering tempreature Tc, in the doped manganites with perovskite structures. The slow magnetic-relaxation in manganese perovskites is interpreted as due to ferromagnetic clusters (solitons), which grow with decreasing temperature down to a temperature at which they freeze due to severe intercluster frustration. Glassy behaviours of manganites are quite unusual and difficult to classify according to existing theories or phenomenology of single-phase materials with random interaction[33]. Matsubara et al.[22] have investigated the ultrafast spin and charge dynamics in the course of a photo-induced phase transition from an insulator with short-range charge order and orbital order to a ferromagnetic metal in perovskite-type GdSrMnO. The magnetization increases with the time constant of 0.5 psec and decay in $\sim$ 10 psec. The interesting point is that the decay time increases with the excitation density. It has been known that the ferromagnetic ordering is due to double-exchange-like interactions of Mn $3d$-e_g state in the perovskite CdSrMnO. It has been suggested that the ferromagnetic interaction induced by the hole seems to be cooperative and non-linear. The size of the magnetic soliton will be determined by competition between the ferromagnetism with the double-exchange process and the antifferomagnetism with the superexchange process. Here we must analyze the quantized magnetic solitons. Thus we shall use the non-perturbed method of topology in gauge fields by G. 't Hooft[34]. In order to argue in the gauge-invariant formula[34-37], we shall introduce the non-linear gauge fields (Yang-Mills fields) A^a_μ, which mediate the effective ferromagnetic interaction induced by the hole. In addition, based on the important idea[23], it has been proposed that the hedgehog-like soliton in three-dimensional system is specified by rigid-body rotation, which is related to gauge fields of SO(4) symmetry for S^3[24,25,35]. Thus it is thought that the non-linear gauge fields A^a_μ introduced by the hole have a local SO(4) symmetry. Then it is assumed that the SO(4) quadruplet fields, A^a_μ, are spontaneously broken around the photo-induced hole through the Anderson-Higgs mechanism, in the perovskite CdSrMnO. The symmetry breaking contains the effects from the electronic frustration and the Jahn-Teller frustration around the doped hole in perovskite CdSrMnO[16,34,37]. We set the symmetry breaking $\langle 0|\phi_a|0\rangle = \langle 0,0,0,\mu\rangle$ of the

Bose field ϕ_a in the Lagrangian density as follows,

$$\begin{aligned} L &= \frac{1}{2}\left(\partial_i S^j - g_1 \varepsilon_{i\hat{j}k} A_i^a S^k\right)^2 \\ &+\frac{1}{2}\left(\partial_i O^j - g_5 \Gamma_{jk}^a A_i^a O^k\right)^2 \\ &+\psi^+\left(i\partial_0 - g_2 T_a A_0^a\right)\psi \\ &-\frac{1}{2m}\psi^+\left(i\nabla - g_2 T_a A_{(\mu\neq 0)}^a\right)\psi \\ &-\frac{1}{4}\left(\partial_\nu A_\mu^a - \partial_\mu A_\nu^a + g_3 \varepsilon_{abc} A_\mu^b A_\nu^c\right)^2 \\ &+\frac{1}{2}\left(\partial_\mu \phi_a - g_4 \varepsilon_{abc} A_\mu^b \phi_c\right)^2 \\ &-\lambda^2\left(\phi_a\phi_a - \mu^2\right)^2 \end{aligned} \tag{1}$$

After the symmetry breaking $\langle 0|\phi_a|0\rangle = \langle 0,0,0,\mu\rangle$, we can obtain the effective Lagrangian density.

$$\begin{aligned} L_{eff} &= \frac{1}{2}\left(\partial_i S^j - g_1 \varepsilon_{i\hat{j}k} A_i^a S^k\right)^2 \\ &+\frac{1}{2}\left(\partial_i O^j - g_5 \Gamma_{jk}^a A_i^a O^k\right)^2 \\ &+\psi^+\left(i\partial_0 - g_2 T_a A_0^a\right)\psi \\ &-\frac{1}{2m}\psi^+\left(i\nabla - g_2 T_a A_{(\mu\neq 0)}^a\right)^2\psi \\ &-\frac{1}{4}\left(\partial_\nu A_\mu^a - \partial_\mu A_\nu^a + g_3 \varepsilon_{abc} A_\mu^b A_\nu^c\right)^2 \\ &+\frac{1}{2}\left(\partial_\mu \phi_a - g_4 \varepsilon_{abc} A_\mu^b \phi_c\right)^2 \\ &+\frac{1}{2}m_1^2\left[(A_\mu^1)^2 + (A_\mu^2)^2 + (A_\mu^3)^2\right] \\ &+m_1\left[A_\mu^1\partial_\mu\phi_2 - A_\mu^2\partial_\mu\phi_1\right] \\ &+m_1\left[A_\mu^2\partial_\mu\phi_3 - A_\mu^3\partial_\mu\phi_2\right] \\ &+m_1\left[A_\mu^3\partial_\mu\phi_1 - A_\mu^1\partial_\mu\phi_3\right] \\ &+g_4 m_1\left\{\phi_4\left[(A_\mu^1)^2 + (A_\mu^2)^2 + (A_\mu^3)^2\right]\right\} \\ &-g_4 m_1\left\{A_\mu^4\left[\phi_1 A_\mu^1 + \phi_2 A_\mu^2 + \phi_3 A_\mu^3\right]\right\} \\ &-\frac{m_2^2}{2}(\phi_4)^2 - \frac{m_2^2 g_4}{2m_1}\phi_4(\phi_a)^2 - \frac{m_2^2 g_4^2}{8m_1^2}(\phi_a\phi_a)^2, \end{aligned} \tag{2}$$

where S^j is the spin of Mn, O^j is the pseudospin operator for the orbital degree of freedom. ψ is the Fermi field of the hole, $m_1 = \mu \cdot g_4$, $m_2 = 2(2)^{1/2}\lambda \cdot \mu$. Here $\hat{j}$ corresponds to the reverse direction of the spin one of the hole. The effective Lagrangian describes three massive gauge fields A_μ^1, A_μ^2, and A_μ^3, and one massless gauge field A_μ^4. The generation function $Z[J]$ for Green functions[38] is shown as follows,

$$Z[J] = \int \mathcal{D}A\mathcal{D}B\mathcal{D}S\mathcal{D}C\mathcal{D}\bar{C}\mathcal{D}\psi^+\mathcal{D}\psi\mathcal{D}\phi$$

$$\cdot \exp i \int \mathrm{d}^4x \left(\mathcal{L}_{eff} + \mathcal{L}_{\mathrm{GF+FP}} + J \cdot \Phi \right), \tag{3}$$

$$\mathcal{L}_{\mathrm{GF+FP}} = B^a \partial^\mu A^a_\mu + \frac{1}{2}\alpha B^a B^a + i\bar{C}^a \partial^\mu \mathcal{D}_\mu C^a, \tag{4}$$

where B^a and C^a are the Nakanishi-Lautrup fields and Faddeev-Popov fictitious fields, respectively.

$$\begin{aligned} J \cdot \Phi &\equiv J^{a\mu} A^a_\mu + J^a_B B^a + J_O O + J_S \cdot S + \bar{J}^a_C \cdot C^a \\ &+ J^a_{\bar{C}} \bar{C}^a + \bar{\eta}\psi + \eta\psi^+ + J^a_\phi \phi_a \end{aligned} \tag{5}$$

Where $J \cdot \Phi$ represents the interaction terms between fields Φ and external sources J. BRS-quartet[26,27] in the present theoretical formula are $(\phi_1, B^1, C^1, \bar{C}^1)$, $(\phi_2, B^2, C^2, \bar{C}^2)$, (ϕ_3, B^3, C^3, C^3), and $(A^4_{L,\mu}, B^4, C^4, \bar{C}^4)$. Where $A^4_{L,\mu}$ is the longitudinal component of A^4_μ. Thus we need these fields for the unitality condition, although these fields are unobservable and fictitious ones. Because masses of A^1_μ, A^2_μ and A^3_μ are created through the Anderson-Higgs mechanism by introducing the hole, the fields A^1_μ, A^2_μ and A^3_μ exist around the hole within the length of $\sim 1/m_1 \equiv R_C$. Where $m_1 = \mu \cdot g_4$ is the mass, which is introduced through the symmetry breaking in eq.(1), of gauge fields A^1_μ, A^2_μ and A^3_μ. From the first term in eq.(2), the spins S of Mn atoms are induced in the ferromagnet state, where the average spin is parallel to $\hat{j}$ direction, within the length of $\sim R_C$ around the hole. That is, the effective Lagrangian represents that the ferromagnetically aligned Mn spins form clusters, in which the hole is trapped, with the radius, $R_C \sim 1/m_1$. In addition Kanazawa[39] has already discussed the creation process of photoholes in manganites. In order to discuss the dynamics of magnetic solitons, we envisage an effective Hamiltonian, H, for the magnetic-soliton, $O(r_{\tilde{i}})$, which is introduced in eq.(2). In order to discuss the spin dynamics and electron hopping, we envisage an effective Hamiltonian, H, for the magnetic-soliton, $O(r_{\tilde{i}})$,

$$\begin{aligned} H &= -J \sum_{<\tilde{i},\tilde{j}>} \cos(\theta_{\tilde{i}\tilde{j}}/2) O(r_{\tilde{i}}) \cdot O(r_{\tilde{j}}) \\ &+ \frac{1}{2} K \sum_{\tilde{i} \neq \tilde{j}} \frac{O(r_{\tilde{i}}) \cdot O(r_{\tilde{j}})}{|r_{\tilde{i}} - r_{\tilde{j}}|} \end{aligned} \tag{6}$$

and the first sum taken only over nearest neighbor(the distance between each magnetic soliton is $\leq 2R_c$) and the second taken over all pair($\tilde{i} \neq \tilde{j}$ means $|r_{\tilde{i}} - r_{\tilde{j}}| \gg 2R_c$) [28]. This second term, which the power law for Long-range pair of interaction between solitons, is derived from the massless U(1) gauge fields A^4_μ. The detailed derivation should be refered to the theory by G. 't Hooft[34]. $S_{\tilde{i}} \equiv \sum_{i \in (4/3)\pi R^3_c} S_i$. That is, $S_{\tilde{i}}$ is the summation of the ferromagnetic spin, S_i, of Mn within $\sim (4/3)\pi R^3_c(\tilde{i})$ around the photo-induced hole at the site $r_{\tilde{i}}$. $S_{\tilde{i}}$ represents the effective spin of the soliton $O(r_{\tilde{i}})$. $\theta_{\tilde{i}\tilde{j}}$ is the angle between $S_{\tilde{i}}$ and $S_{\tilde{j}}$. The first term corresponds to short-range ferromagnetic ordering interaction and the second corresponds to long-range frustration. If g_3 in eq.(1) is assumed to be equal to $(\pi/K)^{1/2}$ [28], where K is the long-range interaction constant in the effective Hamiltonian in eq.(6). Although the first term of the effective Hamiltonian in eq.(6) cannot be derived immediately from the effective Lagrangian in eq.(2), this term can be introduced approximately as follows. When the magnetic soliton,$O(r_{\tilde{i}})$, with the effective spin $S_{\tilde{i}}$ is located in the nearest neighors of the magnetic soliton,$O(r_{\tilde{j}})$, with the effective spin $S_{\tilde{j}}$, holes are hopping between two solitons $O(r_{\tilde{i}})$ and $O(r_{\tilde{j}})$. If $S_{\tilde{i}}$ is parallel to

$S_{\tilde{j}}$, the exchange interaction induces much reduction of the kinetic energy. In order to discuss the correlation among spins $S_{\tilde{i}}$, we shall consider by the effective Hamiltonian with the p-spins interaction term,

$$H_{eff} = -\sum_{\tilde{i_1}<\ldots<\tilde{i_p}}^{N} K_{\tilde{i_1}\ldots\tilde{i_p}} S_{\tilde{i_1}} \ldots S_{\tilde{i_p}} + \sum_{\tilde{i}}^{N} h_{\tilde{i}} S_{\tilde{i}} \tag{7}$$

The spherial spin-constraint is $\sum_{\tilde{i}=1}^{N} S_{\tilde{i}}^2 = lN$, using $\sum_{\tilde{i}=1}^{N}(S_{\tilde{i}}^2 - l) = 0$. The couplings are Gaussian variables with zero mean and average $\overline{K_{\tilde{i_1}\ldots\tilde{i_p}}^2} = \frac{p!}{2(lN)^{p-1}}$. The relaxation dynamics is given by the Langevin equation,

$$\partial_t S_{\tilde{i}}(t) = -\beta \frac{\delta H_{eff}}{\delta S_{\tilde{i}}(t)} - z(t) S_{\tilde{i}}(t) + \eta_{\tilde{i}}(t) \tag{8}$$

β is 1/T. Where $\eta_{\tilde{i}}(t)$ are Gaussian random variable, with zero mean and variance 2. The second term on the right-hand side enforces the spherical constraint. The two-time correlation and the linear response functions are represented as,

$$C(t,t') = \frac{1}{N}\sum_{\tilde{i}=1}^{N} < S_{\tilde{i}}(t) S_{\tilde{i}}(t') > \tag{9}$$

$$R(t,t') = \frac{1}{N}\sum_{\tilde{i}=1}^{N} \frac{\partial < S_{\tilde{i}}(t) >}{\partial h_{\tilde{i}}(t')} \tag{10}$$

The dynamical equations for them can be obtained from eq.(8) through standard function methods[29].

$$\begin{aligned} \partial_t C(t,t') &= -[1 - p\beta\varepsilon(t)] C(t,t') + 2R(t',t) \\ &+ \frac{p\beta^2}{2}\int_0^{t'} dt" C^{p-1}(t,t") R(t',t") \\ &+ \frac{p\beta^2(p-1)}{2}\int_0^{t} dt" R(t,t") C^{p-2}(t,t") \\ &\cdot C(t",t'). \end{aligned} \tag{11}$$

$$\begin{aligned} \partial_t R(t',t) &= -[1 - p\beta\varepsilon(t)] R(t',t) + \delta(t-t') \\ &+ \mu(p-1)\int_{t'}^{t} dt" R(t,t") C^{p-2}(t,t") R(t",t') \end{aligned} \tag{12}$$

$\varepsilon(t)$ can be identified as the energy per spin multiplying eq.(8) by $S_{\tilde{i}}(t')$, averaging over the noise and the couplings and taking the limit $t' \to t$. From the first term in eq.(11) in the condition of $p\beta\varepsilon(t) < 1$, it is seen that the relaxation time is $\propto \frac{1}{1-p\beta\varepsilon(t)}$ approximately. When the intensity of the optical pulse increases, number of hole-induced magnetic solitons increses. As a result, p increases and then the relaxation time increases. This is consistent with the recent experiment[22]. For $\tau \equiv t - t'$ finite and $\tau/t \longrightarrow 0$ asymptotically, time homogeneity and the

fluctuation-dissipation theorem (FDT) hold. For large values of τ, $C_{\rm FDT}(\tau)$ tends to a value q and $R_{\rm FDT}(\tau)$ tends to zero. In the FDT regime[40], eq.(11) yields

$$\left(\frac{\partial}{\partial\tau}+1\right)C_{\rm FDT}(\tau)+(\mu+p\beta\varepsilon_\infty)[1-C_{\rm FDT}(\tau)] = \mu\int_0^\tau d\tau" C_{\rm FDT}^{p-1}(\tau-\tau")\frac{dC_{\rm FDT}(\tau")}{d\tau"} \tag{13}$$

with the asymptotic energy ε_∞ given by

$$\varepsilon_\infty = -\frac{\beta}{2}\left[(1-q^p)+pq^{p-1}\int_0^1 d\lambda" g(\lambda")C^{p-1}(\lambda")\right]. \tag{14}$$

Here $g(\lambda" \equiv t'/t) = tR(t,t')$.
The correlation decays to a value q determined by

$$1-p\beta\varepsilon_\infty+\mu(1-q^{p-1}) = -\frac{1}{1-q}. \tag{15}$$

3. Conclusion

The photo-induced magnetic soliton in the perovskite CdSrMnO has been introduced, by using the theoretical formula, which is on the gauge-invariant effective Lagrangian density. We have discussed the dynamics of photo-induced magnetic solitons. We propose the magnetic relaxation mechanism of photo-induced magnetic solitons in the perovskite CdSrMnO by using the effective Hamiltonian with the p-magnetic solitons interactions and the Langevin equation.

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