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Vortex Patterns in Layered Magnets with Nonmagnetic and Magnetic Impurities

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Effects of a single-atom impurity on a small vortex-like soliton are studied in the two-dimensional (2D) Heisenberg model of layered magnets. In the 41×41 spin lattice with exchange parameters corresponding to the 2D easy-plane copper ferromagnet and the easy-axis manganese antiferromagnet a vortex-like structure and its energy are calculated in the presence of a single nonmagnetic or magnetic impurity. It is found that the in-plane and out-of-plane vortices are attracted to and centered on the impurity in the easy-plane magnet up to the almost isotropic limit. Layered Cu-halide compounds are found to be good candidates for experimental observation of stable, fixed vortex–antivortex patterns.

Introduction Layered compounds are well-known [1] to exhibit quasi-two-dimensional (2D) magnetism at low temperatures. The 2D nature of these structures arises because the ratio of interlayer exchange to intralayer exchange is of the order $J_\perp/J \sim 10^{-3} - 10^{-5}$, and 2D effects can be experimentally observed in a narrow temperature interval (referred to as the fluctuation region) just above the ordering temperature. In this case, the 2D spin lattices can be described by the anisotropic Heisenberg model with the Hamiltonian [1]

$$H = -J \sum_{\mathbf{r},\mathbf{a}} [S^x_{\mathbf{r}} S^x_{\mathbf{r}+\mathbf{a}} + S^y_{\mathbf{r}} S^y_{\mathbf{r}+\mathbf{a}} + \lambda S^z_{\mathbf{r}} S^z_{\mathbf{r}+\mathbf{a}}] - D \sum_{\mathbf{r}} (S^z_{\mathbf{r}})^2 \,, \tag{1}$$

where $\lambda = J^z/J$ characterizes the exchange anisotropy, D is a constant of the single-ion anisotropy, $\mathbf{S_r}$ is a classical spin on the site with the radius-vector $\mathbf{r} = (n, m)$ on the square lattice and the summation with respect to $\mathbf{a}$ is taken over nearest-neighbor sites. Low-dimensional crystals with copper ions ($S = 1/2$) are 2D ferromagnets with only the exchange-type easy-plane anisotropy of the order $\epsilon = 1 - \lambda \sim 10^{-2} - 10^{-3}$ [11]. They are described by Eq. (1) with $J > 0$ and $D = 0$. As an example, it is remarked that K_2CuF_4 has $J/k = 11.2$ K and $\epsilon \simeq 1.2 \times 10^{-2}$. There is also a class of metal- organic compounds with the general formula $(C_nH_{2n+1}NH_3)_2MCl_4$, where M = Cu are also 2D ferromagnets with the exchange $J_{\mathrm{Cu}} \simeq 18$ K and $\epsilon \simeq 3 \times 10^{-3}$. Also of interest is the case M = Mn ($S = 5/2$) which corresponds to almost ideal 2D antiferromagnets with $J_{\mathrm{Mn}} = -5$ K and anisotropy is of the easy-axis single-ion type rather than anisotropy of exchange origin. These compounds are described by Eq. (1) with $\lambda = 1$ and $\alpha \equiv D/J \simeq 1.1 \times 10^{-3}$.

The electron paramagnetic resonance (EPR) in these compounds reveals Arrhenius behavior, $\exp(E/T)$, in the temperature-dependent linewidth in the fluctuation region

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immediately above the ordering [2]. One of possible explanations of this effect is based on the assumption that excitation of magnetic solitons, or vortices, with energy E [2–4] exist in 2D magnets. Thus, the energy of the vortex can be obtained directly from the experimental linewidth data.

To verify the assumption of the vortex contribution to the EPR linewidth it is useful to study the influence of nonmagnetic and magnetic impurities on the structure and energy of vortices. In [5] it has been shown that a nonmagnetic impurity in the finite-size easy-plane ferromagnet increases drastically the stability of the in-plane (P) vortex. The critical value of the parameter λ, when the P vortex loses stability and the out-of-plane (OP) vortex arises, appears to be close to unity, $\lambda_c = 0.957$.

In the present work we investigate numerically the structure of the P and OP vortices in the easy-plane copper ferromagnets with either a nonmagnetic defect (e.g. Zn, Cd) or a "heavy" magnetic impurity (Mn) as well as the effect of a nonmagnetic and "light" magnetic impurity (Cu) on the stability property of the Skyrmion in the easy-axis manganese antiferromagnet. We use the term "Skyrmion" for a vortex of the Belavin-Polyakov type [6] with the energy close to $4\pi JS^2$, which is independent of the excitation size. This is only valid for the classical isotropic magnet, but the structure and energy remains similar for weak easy-axis anisotropy [3]. Note that a vortex in the 2D magnet with the easy-plane anisotropy has the energy which depends on the system size, R_S. In particular, the energy of the P vortex is equal to $E_P^a = \pi JS^2 \ln (R_S/r_0)$, where r_0 is about the lattice constant [7].

Equations for Spins with Impurity To take into account the effect of a nonmagnetic impurity one has to remove a spin and corresponding exchange interactions with its nearest neighbors from the spin Hamiltonian (1). Our additional assumption about the influence of the nonmagnetic impurity includes also an increase of the easy-plane anisotropy (a decrease of the parameter λ) for the exchange interaction of every spin adjacent the empty site with their three neighbors.

In the case of the magnetic impurity a special question remains about the value of the exchange interaction between spins of Cu^{2+} and Mn^{2+} ions. We know only the estimate for the exchange in CuMn spin-glass alloys [8]. It is ferromagnetic with the value of thousands of Kelvins. However, in numerical calculations, because of a different physical situation for the above-mentioned layered magnets, we use the value of the exchange $|J_{\mathrm{Cu-Mn}}|$ from $|J_{\mathrm{Mn}}|$ to $5|J_{\mathrm{Mn}}|$ which is slightly more than J_{Cu}, and both ferromagnetic and antiferromagnetic types of interaction are considered.

Thus, we consider the spin square matrix of the 41×41 size with the impurity site (21, 21). One needs to solve the static Landau-Lifshitz equations

$$\mathbf{S_r} \times \mathbf{F_r} = \mathbf{0}\,, \qquad \mathbf{F_r} = -\frac{\delta H}{\delta \mathbf{S_r}}\,, \tag{2}$$

where $\mathbf{F_r}$ is the effective field for $\mathbf{S_r}$ on the site with coordinate (n, m). Computing code solving the equation is realized as the interation procedure whose details will be published elsewhere [9]. The main idea of finding solutions of static Eqs. (2) is that as it follows from Eq. (2) $\mathbf{S_r}$ is parallel to the effective field $\mathbf{F_r}$, and hence the interation step can be written as

$$\mathbf{S}_\mathbf{r}^{i+1} = S \cdot \mathbf{F}_\mathbf{r}^i / F_\mathbf{r}^i \tag{3}$$

where $F_{\mathbf{r}}^i$ is the length of the vector $\mathbf{F}_{\mathbf{r}}^i$. If one starts with the initial vortex-like spin distribution corresponding to approximate vortex solutions for the easy-plane magnet [7] or to the Skyrmion for the easy-axis anisotropic system [3] then the iteration procedure converges very fast to the vortex–impurity complex solution if the latter is stable. The energy of the vortex–impurity complex is calculated as the difference between the total energy of a system with vortex–impurity structure, E_{tot}, and the ground state energy of the system with an impurity, E_0.

Results and Discussion In this section we present results of solving Eqs. (2), which are summarized as the following.

For the case of a nonmagnetic impurity in the easy-plane ferromagnet we confirm the analytical result of the work [5] that λ_c determining the boundary of the stability of the P vortex is close to the isotropic case. For the 41×41 spin matrix the value appears to be equal to $\lambda_c = 0.953$ which is determined from the magnetization curve. The ground state energy of the system with a nonmagnetic impurity in the unit of JS^2 equals to $E_0 = -3276$. Then the energy of the P vortex centered on the site without a spin is $E_P = 10.61$. It is close to the analytical estimate $E_P^a = 11.67$ for the perfect spin lattice with $R_S = 41$ and $r_0 = 1$.

For $\lambda > \lambda_c$ the OP vortex arises with nonzero out-of-plane z-component. This solution is also stable up to values $\lambda_* = 0.993$. In Fig. 1 the typical structure of the OP solution is presented for $\lambda = 0.988$ corresponding to the parameter of the 2D ferromagnet K_2CuF_4. A smaller length of spin projections on the XY-plane near the impurity corresponds to a larger value of the out-of-plane components. For smaller values of the anisotropy, $\lambda > \lambda_*$, the vortex is not captured by the impurity. In this almost isotropic limit energies of vortices with a different location of their centers (on the site or between sites) is almost equal and the vortex can be shifted from the impurity virtually without a change of its energy. In general the energy of the OP vortex localized on a nonmagnetic impurity is less than the energy of the same vortex in the perfect system. As a function of the parameter λ the energy decreases at first slowly in the interval $[\lambda_c, \lambda_*]$ and then very rapidly decreases to zero. As seen above, the copper metal-organic compounds have the anisotropy with $\lambda = 0.997$ in which case pinning the vortex seems to be impossible. On the other hand, the layered ferromagnet K_2CuF_4 doped with the impurity is evidently appropriate for revealing the vortex–impurity complex.

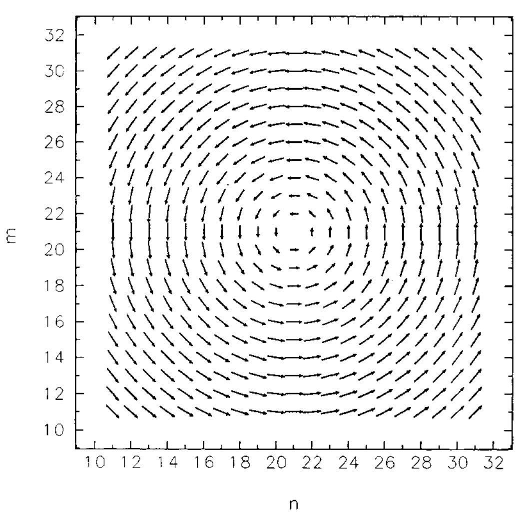

Fig. 1. Out-of-plane vortex centered on the magnetic impurity in the easy-plane ferromagnet ($\lambda = 0.988$). Arrows denote the XY-projections of spins

However, if one assumes that the impurity has an effect on the anisotropic exchange of adjacent spins with their neighbors, then this can result in changing the stability property of the P vortex and critical values of λ. If we assume that the exchange $\lambda' = 0.9$ then we find that λ_c shifts to higher values and becomes equal to 0.962. Hence the onset of the bifurcation of the OP vortex is sensitive to changing interactions near the impurity. In general this could give a possibility to observe the P-vortex–impurity complex in a larger class of layered compounds.

It is important to note that if impurities are arranged in a regular structure then vortice and antivortice are pinned by the impurities and form a periodic pattern. A fragment of the pattern with four impurities and the P vortice and antivortex is shown in Fig. 2.

The effect of a magnetic impurity is qualitatively the same as that of a nonmagnetic impurity. In the OP vortex the spin of the impurity is perpendicular to the easy-plane and does not disturb the symmetry of vortex configuration. In the P vortex the impurity spin lies in the easy plane but the effective field on its site is zero hence the spin direction is arbitrary within the plane unless one considers an additional anisotropy. However, if one needs a variation of the energy of the vortex–impurity complex then the magnetic doping allows this effectively. In fact, the total energy of the system with the P-vortex–impurity complex does not depend on the value of interaction between host and impurity spins. At the same time the ground state energy of the system with the impurity changes explicitly. This leads to the possibility of controlling the variation of energy of the P-vortex–impurity complex.

At last it is be noted that due to the symmetry of the Landau-Lifshitz equations (2) all static solutions for the easy-plane ferromagnets with impurities are transformed into corresponding solutions for the easy-plane antiferromagnets by reversing the direction of the every second spin.

Before we present the data showing the influence of the impurities on the Skyrmion properties it is remarked that they are dynamically unstable in magnets with weak easy-axis single-ion anisotropy [3]. To stabilize the Skyrmion one has to add some definite types of interactions, e.g., the Dzyaloshinski-Moriya interaction [10], which actually occurs in metal-organic compounds. Nevertheless, we have found the static Skyrmion-like solutions in the discrete system with the magnetic impurity has a long lifetime before it loses its stability. Its energy is calculated and equal to $E_S = 12.36$ that is close to 4π. The XY-projection of the solution is presented in Fig. 3. Indeed the existence

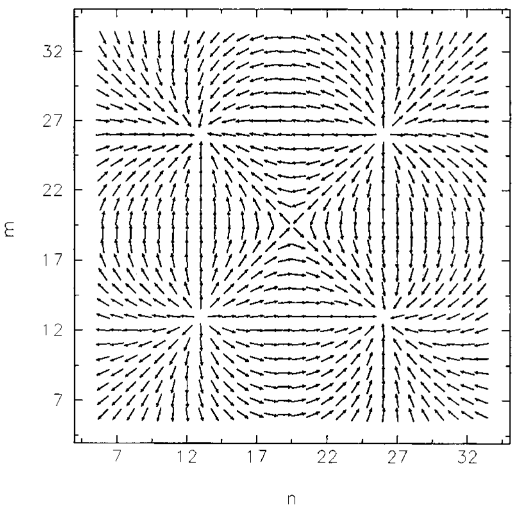

Fig. 2. Vortex–antivortex pattern in the easy-plane ferromagnet with a regular location of nonmagnetic impurities ($\lambda = 0.7$)

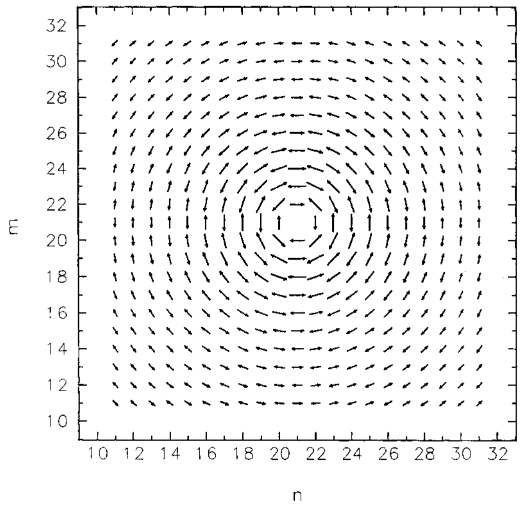

Fig. 3. XY-projections of the Skyrmion-like structure localized on a magnetic impurity in the easy-axis antiferromagnet ($\alpha = 1.1 \times 10^{-3}$)

of the Skyrmion-impurity complex is easily proved at least in the case of the special relation between values of host (Mn) and impurity (Cu) spins and exchange constants: $S_{\rm Mn}/S_{\rm Cu} = |J_{\rm Cu-Mn}/J_{\rm Mn}|$. In this situation the action of the impurity on matrix spins appears to be equivalent to that of the original spin. Then the solution is obtained from the Skyrmion of the pure system by a simple replacement of the host spin by the impurity ion. Thus, in real metal-organic antiferromagnets with weak but complex anisotropies the Skyrmion–impurity complex may be accessible with the energy close to $4\pi JS^2$.

In conclusion, we note that the present consideration shows that the copper easy-plane compounds with magnetic and nonmagnetic impurities are good candidates for experimental realization and observation of stable vortex–antivortex patterns. The EPR study in the crystals would allow to obtain estimates for the vortex energy and parameters of the spin–impurity interaction from analysis of the temperature-dependent linewidth data.

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