Collective mode dynamics of the helical magnets coupled to electric polarization via spin-orbit interaction is studied theoretically. The soft modes associated with the ferroelectricity are not the transverse optical phonons, as expected from the Lyddane-Sachs-Teller relation, but are the spin waves hybridized with the electric polarization. This leads to the Drude-like dielectric function $\varepsilon(\omega)$ in the limit of zero magnetic anisotropy. There are two more low-lying modes; phason of the spiral and rotation of helical plane along the polarization axis. The roles of these soft modes in the neutron scattering and antiferromagnetic resonance are revealed, and a novel experiment to detect the dynamical magneto-electric coupling is proposed.

The gigantic coupling between the magnetism and ferroelectric properties is now an issue of keen interest 
1, 2, 3, 4, 5, 6, 7, 8, 9]. A representative system of interest is $RMnO_3$ with $R=$Gd,Tb,Dy. For example, TbMnO$_3$ shows a ferroelectric moment $P/c$ below a temperature $T_{FE} = 28K (< T_{Neel} = 42)$, and furthermore changes its direction of electric polarization to a axis under the magnetic field $H/b$ 11, 12. Similar strong coupling behavior has been observed in Ni$_3$V$_2$O$_8$ 8, R$Mn_2$O$_5$ ($R=$Tb,Ho,Dy) 13, 14, Ba$_{0.5}$Sr$_{1.5}$Zn$_2$Fe$_{12}$O$_{22}$ 8. A common and essential feature of these compounds is that there are frustrations in the magnetic interactions. For $RMnO_3$, Kimura et al. 2 revealed that the increased GdFeO$_3$-type distortion of perovskite lattice leads to the further-neighbor exchange interactions and nontrivial magnetic structures. Later the neutron scattering experiment determined the spin structure of TbMnO$_3$; it shows the incommensurate collinear spin ordering pointing along b-direction for $T_{FE} < T < T_{Neel}$, while the helical spin structure winding within the $b-c$ plane occurs for $T < T_{FE}$ with the helical wave vector $q//b$ 15. A similar helical structure is also observed in Ni$_3$V$_2$O$_8$ 8. These experiments point to key role of the non-collinear spin configurations such as helical spin structure, which are induced by frustrated exchange interactions, in producing the electric polarization and enhanced magneto-electric coupling.

A microscopic mechanism of the ferroelectricity of magnetic origin has recently been proposed by Katsura et al. 10, which is based on the idea that spin current is induced between the noncollinear spins and hence is the electric moment due to Aharonov-Casher effect 11, 12. This result can be regarded as an inverse effect of Dzyaloshinskii-Moriya interaction 13. Phenomenological treatment of this mechanism 14 and its extension to include electron-lattice interaction 15, have been also reported. By now the static and/or ground state properties of the magneto-electric coupled systems are well understood. The obvious next step is to study their novel dynamic properties searching for the unique electromagnetic effects. In this context, we recall that the collective modes are a central issue in the research on ferroelectrics, where one of the transverse optical (TO) phonons softens toward the phase transition and is condensed according to the Lyddane-Sachs-Teller relation. This conventional view is not relevant in the case of multiferroics since the lattice displacement is not essential to the electronic polarization. Therefore it is an important open problem to identify relevant collective mode that are responsible for ferroelectricity of magnetic origin.

In this paper, we study exactly this question, i.e., the low energy dynamics of the system of the coupled spins and electric polarization. We found the new collective modes of spin and polarization waves that couple the dielectric and magnetic properties in a novel way. Experiments aimed at the detection of these collective modes in terms of the magnetic resonance, neutron scattering and the ac-dielectric measurement are also discussed.

In ref. 10, Katsura et al. have shown that spin (super)current in noncollinear magnets $j_s(\propto \mathbf{S}_i \times \mathbf{S}_j)$ leads...
to the electric polarization $\vec{P} \propto \vec{e}_{ij} \times \vec{j}_i$ with $\vec{e}_{ij}$ being the unit vector connecting two sites $i$ and $j$. An effective Hamiltonian describing this coupling between the spins and the atomic displacement $\vec{u}_n$, which represents the electric polarization including the displacement of electronic cloud, is given as:

$$H = H_1 + H_2 + H_3 + H_4,$$

$$H_1 = -\sum_{m,n} J(R_m - R_n) \vec{S}_m \cdot \vec{S}_n,$$

$$H_2 = -\lambda \sum_m (\vec{u}_m \times \vec{e}_z) \cdot (\vec{S}_m \times \vec{S}_{m+1}),$$

$$H_3 = \sum_m \frac{\kappa}{2} \vec{u}_m^2 + \frac{1}{2M} \vec{P}^2,$$

$$H_4 = \sum_m D(S_m^y)^2.$$

Here, the spin-lattice interaction $\lambda$ stems from the relativistic spin-orbit interaction and corresponds to the DM interaction once the static displacement $\langle \vec{u}_m \rangle$ is non-zero and breaks the inversion symmetry. Note that purely electronic polarization is possible even without the atomic displacement, but the latter is always accompanied with the inversion symmetry breaking. Therefore $\vec{u}_m$ should be regarded as the lowest frequency representative coordinates relevant to the polarization, i.e., the transverse optical phonon, and the polarization $\vec{p}$ is given by $e^* \vec{u}_m$ with a Born charge $e^*$. The term $D(S^y)^2$ with positive $D$ represents the easy-plane spin anisotropy. Here, we have assumed that the ground state spin configuration on the plane perpendicular to the helical wave vector is ferromagnetic and the low-energy excitations on this plane have a usual parabolic dispersion relation, i.e., $\propto |q_1|^2$. Hence we shall focus on the modes along the helical wave vector which is set parallel to the $z$-axis shown in Fig.1. In $H_1$, the summation is taken over all the combinations of $m, n$ and $J(R_m - R_n)$ denotes the Heisenberg interaction between $\vec{S}_m$ and $\vec{S}_n$ at $R_m$ and $R_n$, respectively. First, we determine the classical ground state configuration of spins and lattice distortions. Since we consider the static configuration, the kinetic term in $H_3$ can be negligible. The variation of $H$ with respect to $\vec{u}_m$ gives $\frac{\delta}{\delta \vec{u}_m} = \frac{\kappa}{2} \vec{e}_z \times (\vec{S}_m \times \vec{S}_{m+1})$. Henceforth we assume that spins are on the easy plane, i.e., $zR$-plane and explicitly given by $S^z_m = \cos(Q R_m + \phi)$, $S^x_m = \sin(Q R_m + \phi)$, $S^y_m = 0$. By substituting this spin configuration into the energy $\varepsilon$ per spin is rewritten as $\varepsilon = -S^2 \vec{J}(q)$ with $\vec{J}(q) \equiv \vec{J}(q) + \frac{\lambda^2 S^2}{\kappa} \sin^2(q a)$. Here $\vec{J}(q) = \sum_m \vec{J}(R_m) e^{-iqaR_m}$ is the Fourier transform of $\vec{J}(R_m)$ and $a$ denotes the lattice constant. The wave number $Q$ is determined to maximize $\vec{J}(q)$. Using $Q$, the uniform lattice displacement is given as $\vec{u}_n = -\frac{\lambda S^2}{\kappa} \sin(Q a) \vec{e}_x$, and hence the static electric polarization is given as $\vec{p} = -\vec{e}_z \times \vec{j}$ with $\vec{j} = e^* \vec{u}_m = e^* \frac{\lambda S^2}{\kappa} \sin(Q a)$.

Now we consider the equations of motion for spins and displacements, and study the collective modes of this system. We introduce a rotating local coordinate system $\xi, \eta, \zeta$ such that the $\zeta$-axis coincides with the equilibrium spin direction at each site, the $\xi$-axis is perpendicular to this direction in $zx$-plane and the $\eta$-axis parallel to the $y$-axis (see Fig.1). Assuming the small quantum/thermal fluctuations around the classical configuration, $S^z_n$ and $u^z_n$ can be regarded as a constant and the equation of motion can be written as $\dot{S}^z_n = \frac{\lambda^2 S^2}{\kappa} \sin^2(q a)$ (2)

$$\dot{S}^\xi_n = \sum_m 2 J(R_m - R_n) S^\xi_m \cos(Q(R_m - R_n)) + \frac{\lambda^2 S^2}{\kappa} \sin^2(q a)(S^\xi_{n+1} + S^\xi_{n-1}) + \lambda S (u^y_n \cos(Q R_{n+1} + \phi) - u^y_{n-1} \cos(Q R_{n-1} + \phi)),$$

Here, we regard $S^z, S^\eta, u^y$ as sufficiently small and neglect their second- or higher order terms. We can also write down the equation of motion for $u^y$. Introducing the Fourier components of $S^\xi, S^\eta, u^y_m$ and $p^\eta_m$, the equations of motion are given by

$$\dot{S}^\xi_q = -A(q) S^\xi_q,$$

$$\dot{S}^\xi_q = B(q) S^\eta_q - i\lambda S^2 \left( \frac{e^{iQa} - e^{-iQa}}{2i} e^{i\phi} u_q - Q \rightarrow -Q \right),$$

$$\dot{u}_q = -\frac{\kappa}{M} p_q,$$

$$\dot{p}_q = -\kappa u_q + i\lambda S \left( \frac{e^{iQa} - e^{-iQa}}{2i} e^{i\phi} S^\eta_{-q} + (Q \rightarrow -Q) \right),$$

where

$$A(q) = 2S \left[ J(Q) - J(Q + q) + J(Q - q) \right]$$

$$+ \frac{\lambda^2 S^2}{\kappa} \sin^2(qa/2) \sin^2(Q a),$$

$$B(q) = 2S \left[ J(Q) - J(Q - q) + \frac{\lambda^2 S^2}{\kappa} \sin^2(Q a) + D \right].$$

We note that $A(q)$ and $B(q)$ satisfy the relations $A(-q) = A(q), B(-q) = B(q)$, respectively, and $A(0)$ is equal to zero.

From Eq,(7), one can see the coupling between the spin wave modes and electric polarization. First $S^\xi$ and $S^\eta$ are canonical conjugate variables, and form a harmonic oscillator at each $q$ in the rotated frame. This spin wave at $q$ is coupled with the phonon $u$ at $q \pm Q$, or $u_q$ is coupled to
$S^\alpha /S^\xi$ at $q \pm Q$. Especially the uniform lattice displacement $u_0^0$ is coupled to $e^{-iQ_\alpha x}S^\alpha_Q - e^{iQ_\alpha x}S^\alpha_Q$ ($\alpha = \eta, \xi$), which corresponds to the rotation of both the spin plane and the direction of the polarization along the $z$-axis. This mode is the Goldstone boson with frequency $\omega = 0$ when $D = 0$, i.e., the symmetric case around $z$-axis. On the other hand, $e^{-iQ_\alpha x}S^\alpha_Q + e^{iQ_\alpha x}S^\alpha_Q$ corresponds to the rotation of spin plane along $x$-axis, which is decoupled to the polarization but is gapped by the effective spin anisotropy introduced by the spin-lattice interaction. The spin wave mode at $q = 0$ corresponds to the sliding mode, i.e., phason, of the spiral. This is decoupled from $u$ and has zero energy at $q = 0$.

Using Eq. (4) with canonical commutation relations $[u_q, p_{q'}] = i\delta_{q,-q'}$ and $[S^\xi_q, S^\eta_{q'}] = i\delta_{q,-q'}$, the matrix form of the retarded Green's function

$$G^R(AB; t-t') = -i\theta(t-t')\langle [A(t), B(t')] \rangle$$

(9)

where $\omega_p = \sqrt{A(Q)B(Q)}$ is the frequency of the spin-plane rotation mode along $x$-axis, i.e., $e^{-iQ_\alpha x}S^\alpha_x + e^{iQ_\alpha x}S^\alpha_x$, and $\omega_0 = \sqrt{\kappa/M}$ is that for the original phonon. This response function has the poles at $\omega_{\pm}$, which are explicitly given by

$$\omega_{\pm}^2 = \frac{1}{2} \left( \omega_0^2 + \omega_p^2 \pm \sqrt{\omega_0^2 + \omega_p^2 - 4A(Q)D\omega_0^2} \right)$$

(11)

Assuming $D, \lambda \ll \omega_0^2$, one can estimate as $\omega_\pm \approx \sqrt{A(Q)D} \sim \sqrt{\kappa SJD}$, and $\omega_\pm \approx \omega_0$. With this frequencies, one can write $\varepsilon_{xy}(\omega) = 1 + \sum I_{\pm} \varepsilon_{\omega_{\pm}}$ with $\omega_{\pm}$ being given by $I_{-} = \frac{\omega_p^2 - \omega_0^2}{M \omega_\pm}$, and $I_{+} = \frac{\omega_0^2 - \omega_p^2}{M \omega_\pm}$. Note that the integral $-\int_{-\infty}^{\infty} d\omega \varepsilon_{xy}(\omega)$ is given by $(\pi/2)(I_- + I_+)$. Here, the two modes contributing to the dielectric function are (i) the phonon mode with the frequency $\omega_{\pm} \approx \omega_0$, and (ii) the $z$-axis rotation mode at $\omega_\pm \approx \sqrt{A(Q)D}$. Usually $\omega_\pm$ is a high frequency and does not show any softening in the present case. The ferroelectricity is due to the spin ordering, which is hybridized with the polarization mode. In other words, the low frequency dielectric function is mostly due to the spin wave mode at $\pm Q$.

Note the low-frequency behavior of $\varepsilon_{xy}(\omega)$ is similar to the Drude form when $D = 0$, and the oscillator strength $I_- (\omega = \omega_-)$ is enhanced as $I_- \sim \omega^2 \sqrt{J/D}$ as $D \to 0$. This means that even though the spin-polarization coupling $\lambda$ and hence the static polarization $p$ is small, the spin wave mode can contribute significantly when $D$ is small.

**neutron scattering spectra** — Next, we shall examine how the spin-polarization coupling affects the (inelastic) neutron scattering spectra. The intensity of the neutron scattered with the momentum transfer $q$ is estimated as

$$\frac{d^2\sigma}{d\omega dq} \propto -\text{Im} \sum_{\alpha \in \perp} G^R(S^\alpha_q S^\alpha_q; \omega),$$

where the superscript $\alpha$ indicates the components perpendicular to $q$. In what follows, for the sake of simplicity, we set $D = 0, \phi = 0$. In order to study the neutron scattering spectra, it is needed to rotate the spin coordinates back to the original laboratory system $S^\alpha (a = x, y, z)$. We can easily derive the transformation formula given by

$$G^R(S^\alpha_q S^\alpha_q; \omega) = \frac{1}{4} \sum_{\alpha \in \perp} G^R(S^\alpha_q + p S^\alpha_q - p'; \omega),$$

$$G^R(S^\alpha_q S^\alpha_q; \omega) = G^R(S^\alpha_q S^\alpha_q; \omega),$$

$$G^R(S^\alpha_q S^\alpha_q; \omega) = \frac{1}{4} \sum_{\alpha \in \perp} \text{sgn}(pp')G^R(S^\alpha_{q+p} S^\alpha_{q-p}; \omega).$$

(12)

For example, at the helical wavevector $q = (0, 0, Q)$, one can read the neutron scattering spectra by using the above transformation formula (13) and the list of pole positions and corresponding intensities summarized in Table (14) and (15). Note that the Green's function is written as $G^R(\omega) = \sum a_i/(\omega^2 - \omega_i^2)$ using the intensity $a_i$ and the pole position $\omega_i$. Now 4 modes are expected to contribute to the neutron spectrum (17): (i) phason mode at $\omega = 0$, (ii) $x$-axis rotation mode at $\omega = \omega_p$, (iii) phonon mode with $q = 0 \at \omega = \sqrt{\omega_0^2 + \omega_p^2}$, and (iv) phonon mode with $q = Q \at \omega_q$. The former two, i.e., (i) and (ii), are of magnetic origin, and can be detected without the spin-polarization coupling, while the weights of the latter two, i.e., (iii) and (iv), are borrowed from the spin wave, and hence their relative magnitudes to the former ones are roughly estimated as; (iii)/(i) $\sim (\omega_p/\omega_q)^2$, and (iv)/(i) $\sim \lambda^2/(\kappa J)$.

**antiferromagnetic resonance** — We shall briefly discuss the antiferromagnetic resonance in our system. Here, $D = 0$, and $\phi = 0$ are also assumed. Magnetic resonance experiments pick up uniform magnetic excitations and corresponding Green's functions are $G^R(S^\alpha_0 S^\alpha_0; \omega), (a = x, y, z)$, where axis $a$ corresponds to the direction of applying oscillating magnetic field. Using again the transformation formula (13), one can see the pole of $G^R(S^\alpha_0 S^\alpha_0; \omega)$ occurs at $\omega = \omega_p$ with the intensity $SB(Q)/(4\pi)$, while that of $G^R(S^\alpha_0 S^\alpha_0; \omega)$ at $\omega = \sqrt{A(Q)D}$.
anisotropy induced by the spin-lattice coupling [19]. They estimated the D ∼ Dzyaloshinskii-Moriya coupling is estimated as ...

dielectric response [21]. They observed the peak of polarization mainly consists of the displacement of the 

electrodes and apply the a.c. electric field Eν with frequency ω along y-direction. Then the magnetization of the same frequency ω can be detected at the edges of the sample polarized along z-direction. Now ω ∼ 20cm−1 corresponds to the Tera-Hertz region, and the highest intensity of the electric field there is ∼ 105V/cm. Putting this value and all the other parameters estimated above with overdamping γ ∼ ω ∼ 20cm−1, we obtain the estimation for the magnitude of the a.c. magnetization as mν ∼ 0.4 × 10−4μB/Mn atom, which can be detected by Kerr rotation spectroscopy. We should note here that the magnitude of mν is much enhanced when γ is small.

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