Magnetoelectric coupling in the honeycomb antiferromagnet $Co_4Nb_2O_9$

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The magnetic structure and magnetoelectric effect have been investigated for single crystals of the antiferromagnet $Co_4Nb_2O_9$. Single-crystal neutron diffraction and magnetic susceptibility measurement have revealed that the magnetic structure is different from a collinear arrangement with spin parallel to the trigonal axis as proposed previously. Co^{2+} magnetic moments are found to be almost lying in the basal plane, which lowers the magnetic symmetry to $C2/c'$ with the propagation vector $\boldsymbol{k} = 0$. Associated with the magnetic phase transition, a sharp anomaly in the dielectric constant and displacement current indicate the appearance of the magnetoelectric below Néel temperature with a large coupling constant up to 30 ps/m. The existence of off-diagonal components in a magnetoelectric tensor indicates the formation of ferrotoroidic order in $Co_4Nb_2O_9$. Such a magnetoelectric effect can be ascribed to the reduction of symmetry caused by simple antiferromagnetic order in a honeycomb network. In addition, we observed an intriguing ME response associated with a simple antiferromagnetic ordering on the honeycomb network. The induced electric polarization changes its direction by an angle -2θ upon rotating magnetic field around the trigonal axis by an angle θ. We attribute the variation of electric polarization direction in a rotating magnetic field to the continuous rotation of magnetic moment on the honeycomb. Our findings may open a variety of novel ME responses based on a honeycomb magnet.

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