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Science 302, 92 (2003);
DOI: 10.1126/science.1089408

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which is much larger than the lattice spacing (or the Fermi wavelength). Here, the Mott insulator can be thought of as a state in which holes and doubly occupied sites form bound states due to their Coulomb interaction. The spatial extension $\xi$ of these bound states is related to their energy (the Mott gap $\Delta$) by $\Delta \sim h^2/(2m^* \xi^2)$. Given the measured value of $\Delta$ in samples close to the transition, this leads to the conclusion that $\xi$ is indeed a large length scale, of order a few nanometers. Finally, we emphasize that our results provide experimental support to the early idea of (12) and to recent theories of the Mott critical endpoint based on the DMFT approach (13–15). Although further effort should be devoted to the inclusion of lattice degrees of freedom in these theories, simplified treatments of these effects (16) do emphasize the key role of electronic degrees of freedom in the transition.

References and Notes
17. We acknowledge fruitful discussions with R. Chitra, S. Flens, G. Kotliar, H. R. Krishnamurthy, M. Rozenberg, and A. J. Millis.

Supporting Online Material
www.sciencemag.org/cgi/content/full/302/5642/89/DC1
Figs. S1 to S3
24 June 2003; accepted 20 August 2003

The Anomalous Hall Effect and Magnetic Monopoles in Momentum Space
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Efforts to find the magnetic monopole in real space have been made in cosmic rays and in particle accelerators, but there has not yet been any firm evidence for its existence because of its very heavy mass, $\sim 10^{16}$ giga-electron volts. We show that the magnetic monopole can appear in the crystal momentum space of solids in the accessible low-energy region ($\sim 0.1$ to 1 electron volts) in the context of the anomalous Hall effect. We report experimental results together with first-principles calculations on the ferromagnetic crystal SrRuO3 that provide evidence for the magnetic monopole in real space.

Dirac (1) postulated in 1931 the existence of a magnetic monopole (MM), searching for the symmetry between the electric and magnetic fields in the law of electromagnetism. A singularity in the vector potential is needed for this Dirac MM to exist. Theoretically, the MM was found (2, 3) as the soliton solution to the equation of the non-Abelian gauge theory for grand unification. However, its energy is estimated to be extremely large, $\sim 10^{16}$ GeV, which makes its experimental observation difficult. In contrast to this MM in real space, one can consider its dual space, namely, the crystal momentum ($k$)-space of solids, and the Berry phase connection (4) of Bloch wave functions. This MM in momentum space is closely related to the physical phenomenon of the anomalous Hall effect (AHE) observed in ferromagnetic metals.

The AHE is a phenomenon in which the transverse resistivity ($\rho_{xy}$) in ferromagnets contains a contribution from the magnetization ($M$) in addition to the usual Hall effect. The conventional expression for $\rho_{xy}$ is

$$\rho_{xy} = R_o B + 4n R_s M$$

where $B$ is the magnetic field, $R_o$ is the usual Hall coefficient, and $R_s$ is the anomalous Hall coefficient. This expression implicitly assumes that the additional contribution is proportional to $M$, and it is used as an experimental tool to measure $M$ as a function of temperature. This analysis is extensively used in studies of ferromagnetic semiconductors with dilute magnetic impurities, which are the most promising materials for applications in spintronics (6). However, the mechanism of AHE has long been controversial (7–11). The key issues are whether the effect is intrinsic or extrinsic and how to treat the impurity and phonon scatterings. Karplus and Luttinger (7) were the first to propose the intrinsic mechanism of AHE, in which the matrix elements of the current operators are essential. Other theories (8, 11) attribute the AHE to the impurity scattering modified by the spin-orbit interaction, namely, the skew scattering (8) and/or the side-jump mechanism (11). These extrinsic mechanisms are rather complicated and depend on the details of the impurities as well as on the band structure of the materials. Nevertheless, all these conventional theories (7–11) for the AHE derive Eq. 1, as they are based on the perturbative expansion of the spin-orbit coupling (SOC) $\lambda$ and $M$, i.e., $R_s \propto \lambda$.

Recently, the geometrical meaning of the intrinsic-origin AHE (6) has been recognized (12–15). The transverse conductivity ($\sigma_{xy}$) can be written as the integral of the Berry phase curvature (the gauge field) over the occupied electronic states in crystal momentum space (Eq. 5). The MM corresponds to the source of the gauge field or curvature defined by this Berry phase connection. Therefore, the AHE can be a direct fingerprint of the MM in crystal momentum space. The presence of time-reversal symmetry results in $\sigma_{xy} = 0$ in the dc limit from the generic argument, and the group theoretical condition for the nonzero $\sigma_{xy}$ is equivalent to that of finite ferromagnetic moment (supporting online text). Therefore, ferromagnets are neces-

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necessary to study $\sigma_{xy}$ even though the Berry phase connection is more universal and exists even in nonmagnetic materials.

We show by detailed first-principles band calculations combined with transport, optical, and magnetic measurements that the observed unconventional behavior of the AHE and Kerr rotation in the metallic ferromagnet SrRuO$_3$ is of intrinsic origin and is determined by the existence of a MM in $k$-space. The conventional expression (Eq. 1) is not supported by our experimental data, which show a nonmonotonous temperature dependence that even includes a sign change.

SrRuO$_3$ with perovskite structure is an itinerant (metallic) ferromagnet. Ru$^{4+}$ has four $t_{2g}$ electrons with low spin configurations. The 4$d$ orbitals of SrRuO$_3$ are relatively extended and large in its 4$d$-space gauge field. Stoichiometric SrRuO$_3$ has four orbitals of SrRuO$_3$ are relatively extended and large in its 4$d$-space, i.e.

\begin{equation}
\langle n(\alpha)n(\alpha+\Delta\alpha) \rangle = 1 + \Delta\alpha \langle n(\alpha)\nabla_\alpha n(\alpha) \rangle = \exp[-i\Delta\alpha a_\alpha(\alpha)]
\end{equation}

where the vector potential $a_\alpha(\alpha)$ is defined by $a_\alpha(\alpha) = i\nabla_\alpha n(\alpha)$. Although the concept of the Berry phase has broad applications in physics (17), its relevance to the band structure in solids has been recognized only recently and in limited situations, such as the quantum Hall effect under a strong magnetic field (18) and the calculation of electronic polarization in ferroelectrics (19, 20). In this case, the parameter $\alpha$ is the crystal momentum $k$. For the Bloch wave function $\psi_{nk}(r) = e^{i\mathbf{k}\cdot\mathbf{r}}\psi_{n}(\mathbf{r})$, where $n$ denotes the band index and $\psi_{n}$ is the periodic part, the vector potential for the Berry phase $A_{\alpha}(k)$ is

\begin{equation}
A_{\alpha}(k) = i\langle \psi_{nk}\delta(\mathbf{k}) - \psi_{nk}\rangle
\end{equation}

where $\mu$ is the chemical potential. With this vector potential, the gauge covariant position operator $x_\mu$ for the wave packet made out of the band $n$ is given by $x_\mu = \partial_{\mu} - A_{\alpha}(k)$. Therefore, the commutation relation between $x_\mu$ and $x_\nu$ includes the gauge field $F_{\mu\nu} = \partial_{\mu}A_{\nu} - \partial_{\nu}A_{\mu}$ as

\begin{equation}
[x_\mu, x_\nu] = -iF_{\mu\nu}
\end{equation}

which leads to the additional (anomalous) velocity $-i[x_\mu, V(x)] = -iF_{\mu\nu}\partial V(x)/\partial x_\nu$, being transverse to the electric field $E_\nu = -\partial V(x)/\partial x_\nu$. Therefore, the transverse conductivity $\sigma_{xy}$ is given by the sum of this anomalous velocity over the occupied states as (18)

\begin{equation}
\sigma_{xy} = \sum_{\mathbf{k}} n_{\mathbf{k}}[\epsilon_{\mathbf{k}}(k)]\delta_{\mathbf{k}}(k)
\end{equation}

where $F_{\mathbf{k}}(k)$ is the Fermi energy of $\Psi_{\mathbf{k}}(\mathbf{k})$, $\beta$ is the inverse temperature, and $n_{\mathbf{k}}[\epsilon] = 1/(e^{\beta\epsilon} - 1)$ is the Fermi distribution function. Hence, the behavior of gauge field $F_{\mathbf{k}}(k)$ in $k$-space (21) determines that of $\sigma_{xy}$. One might imagine that it is a slowly varying function of $k$, but that is not the case. Fig. 3B is the calculated result for $F_{\mathbf{k}}(k)$ in the real system SrRuO$_3$. It has a very sharp peak near the $\Gamma$-point and ridges along the diagonals. The origin for this sharp structure is the (near) degeneracy and/or the band crossing, which act as MMs. Consider the general case where the two-band Hamiltonian matrix $H(k)$ at $k$ can be written as $H(k) = M(k)t e^{i\theta(k)}$;}

![Fig. 1. Measured temperature dependence of the (A) magnetization $M$, (B) longitudinal resistivity $\rho_{xx}$, and (C) transverse resistivity $\rho_{xy}$ for the single-crystal and thin-film SrRuO$_3$ as well as for the Ca-doped Sr$_{0.8}$Ca$_{0.2}$RuO$_3$ thin film. $\mu_0$, Bohr magneton. (D) The corresponding transverse conductivity $\sigma_{xy}$ is shown as a function of $M$, together with the results of first-principles calculations for cubic and orthorhombic structures (25). In our calculations, the change of magnetization is taken into account by the rigid splitting of up and down spin bands. As $\sigma_{xy}$ should vanish with $M$ at high temperatures, the calculated $\sigma_{xy}$ is multiplied by the additional $M/M_0$ (where $M_0 = 1.5\mu_0$) factor, which does not affect its behavior except in the vicinity of $T_c$.](http://www.sciencemag.org/science/vol302/issue302)
We studied the behavior of $\sigma_{xy}$ by first-principles calculations (16). The calculated density of states is not so different between the cases with and without SOC (Fig. 4A), whereas the $\sigma_{xy}$ should be very sensitive to the Bloch wave functions and depends on the Fermi-level position and the spin-splitting (magnetization) substantially, as predicted by the discussion above. We determined the behavior of $\sigma_{xy}$ as a function of the Fermi-level position by using a small broadening parameter for the lifetime $\delta$ (70 meV) (Fig. 4B). When the Fermi level was shifted, not only the absolute value but also the sign of $\sigma_{xy}$ was found to change. The sharp and spiky structures are the natural results of the singular behavior of the MM (Fig. 3). For the case without any shift of Fermi level, we obtained a value of $\sigma_{xy} = -60 \, \Omega^{-1} \, \text{cm}^{-1}$, which has the same sign as and is comparable with the experimental value (about $-100 \, \Omega^{-1} \, \text{cm}^{-1}$). Such a spiky behavior should also be reflected in the $\sigma_{xy}$-dependent, especially for the low-energy range with longer lifetime, whereas it should be suppressed at higher activation energies with shorter lifetime. As shown in Fig. 2 for the dependence of optical conductivity, the high-energy ($>0.5$ eV) part, which is dominated by the $p - d$ charge transfer peak, is usual and can be well reproduced by our calculations, whereas the observed peak structure of $\sigma_{xy}(\omega)$ below 0.5 eV is a clear demonstration of the predicted spiky behavior. The spectra below 0.2 eV were not measured because of the technical difficulty, but structure there should be even sharper, because the dc limit $\text{Re}(\sigma_{xy}) \approx -100 \, \Omega^{-1} \, \text{cm}^{-1}$ has the opposite sign [the $\text{Im}(\sigma_{xy})$ at the dc limit should go back to zero]. Such low-energy behavior is well represented by our calculations, providing further evidence for the existence of MMs.

It is straightforward to understand the results of our transport measurement for $\sigma_{xy}$. We attribute the temperature ($T$) dependence of $\sigma_{xy}$ to that of the magnetization $M(MT)$. As the result of $k$-space integration over occupied states, the calculated $\sigma_{xy}$ is nonmonotonous as a function of $M$ (Fig. 1D). With the reduction of spin-splitting, the calculated $\sigma_{xy}$ after the initial increase, decreases sharply, then increases and changes sign (becoming positive), and finally decreases again, capturing the basic features of

$$\Sigma_\mu = \sum_{k} f_\mu(k) \sigma_\mu$$
where $\sigma_{1,2,3}$ are the Pauli matrices and $\sigma_0$ is the unit matrix. When $k$ is mapped to the vector $f(k) = [f_1(k), f_2(k), f_3(k)] = f(k) [\cos k, \sin k, \sin k]$ (cos $\theta$, sin $\theta$, sin $\theta$), the contribution to $\sigma_{xy}$ from the neighborhood of this degeneracy region can be given by the $f$-space solid angle $d\Omega_f = \delta(k, \theta) \delta(k, \phi) \, \sin k \, dk_\theta \, dk_\phi = \delta k_\phi \sin \theta \, dk_\theta$ for the infinitesimal $dk_\theta \, dk_\phi$ integrated over $k$. The solid angle corresponds to the flux from the monopole at $k = 0$. The $\sigma_{xy}$ calculated from $f_\mu$ in the $\theta, \phi$ space for $t_{yz}$ bands as a function of $(k_x, k_y)$ with $k_x$ being fixed at 0 for SrRuO$_3$ with cubic structure. The sharp peak around $k_x = k_y = 0$ and the ridges along $k_x = \pm k_y$ are due to the near degeneracy of $d_{yz}$ and $d_{xz}$ bands because of symmetry (supporting online text).
the experimental results. Even more surprisingly, when the measured $p_{xy}$ versus $T$ curves shown in Fig. 1C are converted into the $\sigma_{xy}$ versus $M$ curves shown in Fig. 1D, they now all follow the same trend and match with our calculations. The curves are measured for different samples (with different saturation moments), but all follow the same rule qualitatively and could be simply explained by the reduction of $M$ (22) (Fig. 1A). However, the comparison between the experiments and the calculations should be semi-quantitative, because the results are sensitive to the lattice structures. The calculated $\sigma_{xy}$ for the fictitious cubic structure shows a strong deviation from that obtained for orthorhombic structure, and it changes the sign to be positive at low temperature (at large $M$). Therefore, more accurate information on the structure is needed to obtain the quantitative result. However, such a sensitivity does not affect our main results, i.e., the nonmonotonous behavior of $\sigma_{xy}$. Even the calculations for cubic structure show such behavior and may be used as a guide of possible deviation.

The results and analysis presented here should stimulate and urge the reconsideration of the electronic states in magnetic materials from a very fundamental viewpoint. For example, the MM is accompanied by the singularity of the gauge potential, i.e., the Dirac string (23). As shown by Wu and Yang (23) this means that more than two overlapping regions have to be introduced, in each of which the gauge of the wave function is defined smoothly. This means that one cannot define the phase of the Bloch wave functions in a single-gauge choice when the MM is present in the crystal momentum space. This leads to some nontrivial consequences, such as the vortex in the superconducting order parameter as a function of $k$ (24, 25), and many others are left for future studies.

References and Notes
5. It has been recognized in the original paper by Berry (25) that the degeneracy points in the parameter space acts as a MM where the gauge field is enhanced.

16. Materials and methods are available as supporting material on Science Online.
21. This gauge field is distinct from that of the magnetic field $\cal B$ in real space, although they are analogous to each other. In the presence of $\cal B$, the covariant momentum operator $\cal p$ is given by $\cal p = -i \hbar \cal A + i \hbar \cal E$, where $\cal B = - \nabla \times A$. This leads to the commutation relation $[\cal p_i, \cal p_j] = -i \hbar \delta_{ij}$, etc., and to the Lorenz force due to the magnetic field $\cal B$. Therefore, these two gauge fields, $\cal B$ and $\cal B$, are dual to each other, and the presence of the one does not necessarily mean that of the other.
22. More Ca-doped samples with different concentrations have been measured. They all follow the same trend and are not shown here because of space limitations.

Supporting Online Material
www.sciencemag.org/cgi/content/full/302/5642/92/DC1 Materials and Methods
SOM Text
References and Notes
21 July 2003; accepted 28 August 2003

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Coherent Soft X-ray Generation in the Water Window with Quasi–Phase Matching

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We demonstrate enhanced generation of coherent light in the “water window” region of the soft x-ray spectrum at 4.4 nanometers, using quasi–phase-matched frequency conversion of ultrafast laser pulses. By periodically modulating the diameter of a gas-filled hollow waveguide, the phase mismatch normally present between the laser light and the generated soft x-ray light can be partially compensated. This makes it possible to use neon gas as the nonlinear medium to coherently convert light up to the water window, illustrating that techniques of nonlinear optics can be applied effectively in the soft x-ray region of the spectrum. These results advance the prospects for compact coherent soft x-ray sources for applications in biophysics and in chemical spectroscopy.

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Fig. 4. The calculated (A) density of states (DOS) and (B) $\sigma_{xy}$ as functions of Fermi-level position for the orthorhombic structure of single-crystal SrRuO$_3$. The Fermi level is shifted rigidly relative to the converged solution, which is specified as the zero point here. The sharp and spiky structure of $\sigma_{xy}$ demonstrates the singular behavior of MMs, f.u., the formula unit SrRuO$_3$.

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