I. INTRODUCTION

Setting itinerant electrons with strong spin-orbit coupling on geometrically frustrated lattices offers new possibilities for strongly correlated electron states. Recent attention has focused on the pyrochlore iridates, which are rare earths, for which several theoretical papers propose the existence of topologically nontrivial ground states. Testing such predictions requires advanced experiments, for example, measurements that can reveal unconventional quantum phase transitions. Here, we report an unusual insulator to metal transition in the pressure-temperature phase diagram of the pyrochlore iridate Eu$_2$Ir$_2$O$_7$.

Among pyrochlore oxides, iridates have their $M$ sites occupied by Ir$^{4+}$ ions whose extended 5$d$ orbitals are prone to strong electron-lattice and spin-orbit couplings. Compared with the less extended orbitals of 3$d$ transition-metal oxides, iridates are naively expected to be less strongly correlated. Nevertheless, the strong spin-orbit and electron-lattice couplings can lift the orbital degeneracy of iridium 5$d$ electrons and narrow their bandwidths. Hence, iridates can be delicately poised near a bandwidth-controlled metal-insulator transition (MIT).

Successive replacement of the $R$ site of R$_2$Ir$_2$O$_7$ with larger rare-earth atoms causes a change from insulating to metallic behavior. Progressing from right to left in the lanthanide series, (Lu, Yb, ... , Gd)$_2$Ir$_2$O$_7$ are all insulators; (Eu, Sm, and Nd)$_2$Ir$_2$O$_7$ are on the boundary, looking metallic at high temperatures but insulating at low temperatures, and Pr$_2$Ir$_2$O$_7$ is metallic down to the lowest temperatures. Increasing the $R$ size in the R$_2$Ir$_2$O$_7$ series makes the Ir-O-Ir bond angle wider and the Ir-O bond length shorter. As a result, the iridium $t_{2g}$ bandwidth decreases and eventually passes the metallization threshold beyond a certain $R$ ionic radius.

As the $R$ size increases from Eu to Nd, (Eu, Sm, Nd)$_2$Ir$_2$O$_7$ show several signatures of a weakening low-temperature insulating phase and a strengthening high-temperature metallic phase: (1) the metal-insulator transition temperature $T_{MI}$ is significantly smaller in the Nd compound (36 K) compared to the Eu and Sm compounds (120 and 117 K, respectively); (2) the small and temperature-dependent gap ($\Delta < 10$ meV) of the insulating phase is smallest in the Nd compound; (3) the Nd compound has the smallest inverse $RRR$ ratio defined as $R_{300 K}/R_{30 K}$; and (4) in the high-temperature metallic phase, only the resistivity of the Nd compound is “metallic,” having a positive slope for $\rho(T)$; in Eu and Sm, $\rho(T)$ is “nonmetallic” with a weakly negative slope. (By nonmetallic, we mean a resistivity that rises with decreasing temperature but with power-law temperature dependence as opposed to the exponential temperature dependence of a gapped insulator.)

These three systems are of particular interest at present because it is believed that states near the metal-insulator boundary in the pyrochlore iridates may have a topological nature. In particular, Wan et al. and Witzczak-Krempa et al. predict that a topological semimetallic state separates the insulating and the metallic ground states. This “Weyl semimetal” is a three-dimensional analog of graphene with the chemical potential pinned to Dirac points of chiral states. In order to form, the Weyl semimetallic state requires magnetic order, and due to the vanishing density of states at the chemical potential, the electrical conductivity of the bulk is predicted to vanish when $T \to 0$ K. Electron interactions and correlations of pyrochlore iridates can be tuned by either chemical or physical pressure. For investigating the nature of the low-temperature state, physical pressure has the obvious advantage that it can be tuned continuously. (Continuous substitution studies in the pyrochlore iridates are not useful, since the ground state is sensitive to disorder.) Physical pressure is somewhat different from chemical pressure, however. Increasing the $R$ atomic size increases the Ir-O-Ir bond angle and the lattice parameter in parallel, whereas, hydrostatic pressure increases the former but decreases the latter. Thus, we expect different insights from the application of physical pressure to explore the boundary between metallic and insulating ground states in pyrochlore iridates.
II. EXPERIMENT

Eu₂Ir₂O₇ single crystals were grown at the Institute for Solid State Physics using the potassium fluoride-flux method.¹¹ We pressurized samples measuring approximately 150 × 100 × 30 μm³ in a moissanite anvil cell¹² and measured resistivity as a function of temperature at several pressures in the range of P = 2–12 GPa using a four-terminal ac method. The pressure medium was 7373 Daphne oil, and pressure was monitored by ruby fluorescence spectroscopy at room temperature. A 1-K dipping probe was used in the temperature range of T = 300 to 2 K. Resistivity below 2 K and magnetoresistance (MR) at 10.01 GPa were measured in a dilution refrigerator.

III. RESULTS

Our resistivity data, from 300 to 2 K at nine different pressures from 2.06 to 12.15 GPa, are presented in Fig. 1. The quantitative effects of increasing the pressure from 2 to 12 GPa are dramatic: The room-temperature resistivity falls by a factor of 60, whereas, the 2-K resistivity falls by a factor of 500. Qualitatively, the slope of the resistivity ∂ρ(1)/∂T for T > 100 K changes from negative (nonmetallic) at low pressure to positive (metallic) at 10 and 12 GPa. At lower temperatures, in contrast, the slope of the resistivity is negative at all pressures, but it is nearly 1000 times larger at 2.06 GPa than at 12.15 GPa. We elaborate on the low-temperature resistivity in the Discussion and show that the low-pressure curves have a temperature-dependent gap that closes between 6 and 8 GPa.

The metal-insulator transition, which occurs at 120 K at ambient pressure, does not show up clearly in the raw resistivity. This is also the case at ambient pressure.⁸ However, Fig. 2 shows that, for low pressures, the slope of ln[ρ(T)] vs T changes near 100 K. This change appears to be quite abrupt in

The 3.49-GPa and the 4.61-GPa curves, both in Fig. 2(a) and in the raw data [Fig. 1(b)]. This change in slope is more clearly seen in plots of ∂ρ/∂T vs T [Fig. 2(b)]: For all pressures below 7.88 GPa, ∂ρ/∂T begins to decrease rapidly, with a sharp well-defined onset, near 100 K. We have used this to identify TMI: Down to TMI, the slope of ∂ρ/∂T is roughly constant, then at TMI, it begins to fall rapidly. The inset of Fig. 2(b) shows a clear example at 3.49 GPa. The red arrows in the first two figures correspond to TMI, assigned to the midpoint between where the slope first starts to turn downward and the point where the extrapolated high- and low-temperature slopes meet. Error bars on our phase diagram, discussed below, extend to these two temperatures. The lowest pressure ∂ρ/∂T curves are rather noisy, perhaps because the contact resistances, which improve as the pressure increases, are rather large, but even in these cases, a sharp change can be identified quite accurately.

At 6.06 GPa and above, there is a minimum in ρ(T), and like TMI, it is close to 100 K. The way this minimum develops is shown in Fig. 3. In the lower-pressure curves, the resistivity has a negative slope at all temperatures, but in the 6.06- and 7.88-GPa curves, an intermediate region of ∂ρ/∂T > 0 develops below a local maximum at T* and a local minimum at Tmin. At higher pressures, the maximum has apparently moved above room temperature, so ρ(T) has a positive metallic slope from Tmin to 293 K. The T* crossover occurs at 180 and 270 K on the P = 6.06- and 7.88-GPa curves, respectively.

We have used the qualitatively different resistivity behaviors to construct the phase diagram shown in Fig. 4. The phase diagram can be viewed as four quadrants, corresponding to four distinct regimes of electronic transport. In the

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**Fig. 1.** (Color online) Resistivity as a function of temperature from P = 2.06 to 12.15 GPa. The left-hand panel contains all of our results, showing that the resistivity is strongly suppressed by increasing pressure. The right-hand panel focuses on the intermediate-pressure data. Red arrows indicate the metal-insulator transition (see Fig. 2 and the text); blue arrows indicate the minimum in ρ(T).

**Fig. 2.** (Color online) Locating the metal-insulator transition TMI. The color coding is the same as in Fig. 1. The transition does not have a strong signature in ρ(T), however, panel (a) shows that there is a marked change in slope of ln(ρ) vs T near 100 K at all pressures, while panel (b) shows that the onset of this behavior can be found, for all pressures below 7.88 GPa, in a sharp downturn in ∂ρ/∂T. As a guide to the eye, we have shown plausible extrapolations of the high-temperature slope and the slope immediately below 100 K. At each pressure, we have placed TMI at the midpoint between the onset of the downturn and the temperature at which these extrapolations meet; whereas, the error bars (see Fig. 4) extend to these two temperatures. The inset of (b) zooms in on ∂ρ/∂T near 100 K for 3.49 GPa. In (a), some curves are offset vertically for clarity.
top-left quadrant ($P \lesssim 6$ GPa, $T \gtrsim 100$ K), the system is an “incoherent metal” characterized by a high resistivity and a power-law temperature dependence with a nonmetallic slope, $\partial \rho(T)/\partial T < 0$. The MIT at $T_{\text{MI}}$ separates the incoherent metallic phase from the “insulating” phase in the bottom-left quadrant ($P \lesssim 6$ GPa, $T \lesssim 100$ K), characterized by an exponentially activated resistivity as discussed below.

In the top-right quadrant ($P \gtrsim 6$ GPa, $T \gtrsim 100$ K), the system is a “conventional metal” characterized by a metallic slope $\partial \rho(T)/\partial T > 0$. The crossover at $T_{\text{min}}$ separates the conventional metal and the “negative $\partial \rho(T)/\partial T$ metal” ($P \gtrsim 6$ GPa, $T \lesssim 100$ K), whose resistivity increases with decreasing temperature in a non-Fermi liquid (NFL) power-law fashion [Fig. 3(b)].

The two high-temperature regimes, the conventional metal, and the incoherent metal, are separated by the $T^*$ crossover, at which the slope of $\rho(T)$ changes from positive to negative.

We distinguish between the incoherent metal (in the top-left quadrant) and the negative $\partial \rho/\partial T$ metal (in the bottom-right quadrant). Although they both have power-law dependence on $T$ with negative $\partial \rho/\partial T$, their absolute resistivities differ by a factor of up to 500, and the latter probably has Landau-type quasiparticles as established by the metallic resistivity at high temperatures, whereas, in the incoherent metallic phase, it is likely that quasiparticles have not formed.

This phase diagram is in broad agreement with the effects of chemical pressure\(^8\) but with the clear difference that $T_{\text{MI}}$ changes rapidly with chemical pressure, falling from 201 to 36 K in going from Eu$_2$Ir$_2$O$_7$ to Nd$_2$Ir$_2$O$_7$, whereas, in our measurements, it is nearly pressure independent. Moreover, $T^*$ is not seen in the chemical pressure measurements.

Our phase diagram suggests a connection between the high- and the low-temperature phases of Eu$_2$Ir$_2$O$_7$: The incoherent metal becomes insulating below $T_{\text{MI}}$; the conventional metal crosses over to the negative $\partial \rho/\partial T$ metal below $T_{\text{min}}$; in other words, the transition between the insulating and the negative $\partial \rho/\partial T$ metallic ground states at $P_c = 6.06 \pm 0.60$ GPa coincides with the incoherent-coherent crossover at $T^* > 100$ K.

IV. DISCUSSION

To discuss our results, we start with the high-temperature part of the phase diagram. The incoherent metallic phase is characterized by a high resistivity and a negative $\partial \rho(T)/\partial T$ that are both gradually suppressed by increasing pressure. Metallic phases in the vicinity of localization transitions are usually subject to strong fluctuations in the spin, charge, and orbital degrees of freedom, resulting in unconventional transport properties. The negative $\rho(T)$ slope in the incoherent metallic regime of Eu$_2$Ir$_2$O$_7$ is probably an example of such physics.

The boundary between the two high-temperature regimes is marked by the dashed $T^*$ line in Fig. 4. The broad peaks at $T^*$ (Fig. 3) mark a coherent-incoherent crossover of the quasiparticle dynamics that is also typical of correlated oxides in proximity to a Mott transition.\(^{13,14}\) The $T^*$ crossover has not been observed in the previous chemical pressure measurements in pyrochlore iridates by replacing the $R$ site with larger atoms: It presumably takes place somewhere between Sm$_2$Ir$_2$O$_7$ and Nd$_2$Ir$_2$O$_7$. Moreover, it probably cannot be realized by alloying on the $R$ site, i.e., partially replacing Eu or Sm with Nd because of the extreme sensitivity of these systems to disorder.\(^1\) Physical pressure is, therefore, the only means by which $T^*$ can be observed.

A similar pressure-induced coherent-incoherent crossover has been observed in Gd$_2$Mo$_2$O$_7$, which is located at the boundary between a ferromagnetic (FM) metal and a spin-glass Mott insulator in the $R_2$Mo$_2$O$_7$ series.\(^{15}\) Gd$_2$Mo$_2$O$_7$ goes through a continuous bandwidth-controlled insulator to metal quantum phase transition at $P_c = 2.4$ GPa.\(^{16}\) Simultaneously, a crossover appears at $T^* \approx 150$ K in the resistivity data. $T^*$ shows a strong pressure dependence in both Eu$_2$Ir$_2$O$_7$ (Fig. 4)
and Gd$_2$Mo$_2$O$_7$.\cite{15} The $T^*$ shifts at a rate of $\Delta T^*/\Delta P \sim 50$ K/GPa in Eu$_2$Ir$_2$O$_7$ and $\sim 35$ K/GPa in Gd$_2$Mo$_2$O$_7$.

Turning now to the low-temperature part of the phase diagram, previous papers\cite{8} have treated the low-temperature regimes of Eu$_2$Ir$_2$O$_7$, Sm$_2$Ir$_2$O$_7$, and Nd$_2$Ir$_2$O$_7$ as a Mott-insulating phase with a so-called temperature-dependent gap (a gap that gets smaller with decreasing $T$ with a nondivergent resistivity in the $T \to 0$ K limit). In all three systems, there is a magnetic anomaly at the metal-insulator transition.\cite{8,17,18} Unlike the Sm and Nd systems, however, Eu$_2$Ir$_2$O$_7$ does not show a sharp metal-insulator transition in the resistivity, rather there is a rapid crossover from power-law resistivity at high temperatures to thermally activated behavior below the 120-K magnetic transition. We do not yet have magnetic data at high pressure, which would allow us to unambiguously determine $T_{MI}$, however, the clear signatures in $\partial \rho/\partial T$ and in $\ln(\rho)$ vs $T$ (Fig. 2), which connect smoothly to 120 K at ambient pressure (Fig. 4), give us confidence that we can identify $T_{MI}$.

This is further reinforced by plots of $\ln(\rho)$ vs $1/T$ in Fig. 5(a), which have a straight-line dependence for a significant range of temperatures below $T_{MI}$ as expected for a gapped system. If we plot the gap extracted from this straight-line behavior as a function of pressure, then the gap appears to vanish continuously between 6 and 8 GPa [Fig. 5(b)]. Similarly, if we plot the $T = 0$ K conductivity $\sigma(0)$, obtained by extrapolating our resistivity curves to 0 K (Fig. 6), the conductivity appears to rise continuously across thispressure range. These behaviors are consistent with a Mott-insulating ground state at low pressure followed by a continuous insulator to metal transition between 6 and 8 GPa. In this scenario, the nondivergence of the resistivity as $T \to 0$ K may be due to bulk impurity states. Indeed, $\rho(0)$ shows a strong sample dependence, consistent with impurity states in the bulk. However, the role of disorder in the insulating phase is not clear, and it should be noted that the MIT at $T_{MI}$ cannot be simple disorder-driven Anderson localization because disorder wipes out the insulating phase and leaves the system metallic at all temperatures.\cite{8} Given the presence of spin-1/2 moments on the iridium sites, frustration-induced localization\cite{19} may play a role in the insulating state, whereas, the recent revelation of a commensurate antiferromagnetic order in Eu$_2$Ir$_2$O$_7$ below 120 K from $\mu$SR measurements raises the possibility of gapping of the Fermi surface by a Slater transition.\cite{17}

A difficulty with the simple Mott-insulator interpretation of our phase diagram is that, although the gap closes continuously with pressure, $T_{MI}$ does not vanish, indeed, it is barely affected by pressure (Figs. 4 and 5). It is, therefore, interesting that recent theoretical proposals, based on band-structure and many-body calculations, have variously proposed strong topological insulator ground states,\cite{20,21} and Weyl semimetallic ground states on the boundary of the metal-insulator transition\cite{22-24} in pyrochlore iridates. These suggestions are consistent with our results. First, the finite resistivity at $T \to 0$ K could arise from small intrinsic Fermi pockets or surface states characteristic of topological insulators. Moreover, Hosur et al.\cite{4} show calculated resistivity for the Weyl semimetal that agrees qualitatively with our data. Indeed, the fact that $T_{MI}$ is not affected by pressure, while the gap vanishes and residual conductivity rises continuously across the critical pressure, could be explained if $T_{MI}$ is produced by a magnetic transition of the itinerant Ir 5$d$ electrons (note that, according to crystal-field calculations, Eu has no magnetic moment in this material) that is only weakly pressure dependent, whereas, the underlying electronic structure undergoes (for example) a Lifshitz transition.

We note that our finding of a roughly pressure-independent $T_{MI}$ is in contrast to a recent pressure study on Nd$_2$Ir$_2$O$_7$ (Ref. 22), which observed a monotonic suppression of $T_{MI}$ with increasing pressure. We do not have an explanation for this discrepancy. Aside from the different materials, the only obvious difference between the measurements is that the Nd$_2$Ir$_2$O$_7$ study used NaCl as the pressure medium, which could produce anisotropic pressures, which can have unpredictable effects on geometrically frustrated systems. (In this respect, Daphne oil, which we used, is an improvement on NaCl but still is not ideal at high pressures.)

After the collapse of the insulating phase above 6.06 GPa ($\Delta = 0$, $\sigma(0) \neq 0$), a low-temperature NFL power-law rise in resistivity survives up to the highest pressure in our experiment in the negative $\partial \rho/\partial T$ metal region in the bottom-right quadrant of our phase diagram. The nature of this phase is not clear. Figure 7(a) is a semilogarithmic plot of $\rho(T)$ as a function of pressure, indicates that the pressure-induced insulator to metal quantum phase transition is continuous.
moments in Eu$_2$Ir$_2$O$_7$ (Ref. 24) would seem to rule out a Kondo-like friction.

At high fields, MR tends toward saturation. Similarly, a recent theoretical paper$^{25}$ that ascribes the resistive upturn in Pr$_2$Ir$_2$O$_7$ and in Nd$_2$Ir$_2$O$_7$ at high pressure$^{22}$ to the low-field data at $T = 200$ mK. At high fields, MR tends toward saturation.

At $P = 10.01$ GPa from $T = 300$ K to 100 mK. The NFL resistivity behavior fits to $\rho(T) = \rho(0) - AT^x$ with $x = 1.0 \pm 0.1$ below 20 K for $P > 6$ GPa. While the resistivity upturn of the metallic phase below $T_{\text{min}}$ looks Kondo-like, similar to what is observed in the frustrated Kondo lattice of Pr$_2$Ir$_2$O$_7$, crystall-field analyses that predict no local $f$ moments in Eu$_2$Ir$_2$O$_7$ (Ref. 24) would seem to rule out a Kondo effect. Similarly, a recent theoretical paper$^{25}$ that ascribes the resistive upturn in Pr$_2$Ir$_2$O$_7$ and in Nd$_2$Ir$_2$O$_7$ at high pressure$^{22}$ to frustrated spin-ice-like correlations among the local $f$ electrons, would also appear to be ruled out by the absence of $f$ moments on the Eu sites.

As well as being a common feature of the metallic pyrochlore iridates, a rising resistivity as $T \to 0$ K has also been observed in metallic pyrochlore molybdates at high pressures. Hydrostatic pressure destroys the FM order in the metallic ground states of (Nd and Sm)$_2$Mo$_2$O$_7$ by tuning the relative strength of double and superexchange interactions among Mo d electrons. The resulting order-disorder transition coincides with a change in the resistivity behavior, from conventional metallic to a “diffusive” NFL state,$^{26}$ characterized by a large, relatively weakly $T$-dependent resistivity, not unlike the resistivity of our $\partial \rho / \partial T > 0$ metal and having an upturn in the resistivity as $T \to 0$ K. In this context, it seems significant that the tetragonal antiferromagnet Ba$_2$IrO$_4$ (Ref. 27) has recently been shown to have a continuous pressure-induced insulator to metal transition that is related to suppression of magnetism, but in this case, the low-temperature resistivity at high pressure is metallic. Thus, the upturn in $\rho(T)$ as $T \to 0$ K is not a generic feature of iridates near the boundary of a Mott-insulating ground state, rather the pyrochlore lattice structure seems to play a decisive role.

To further investigate the negative $\partial \rho / \partial T$ metallic phase, we measured MR at $P = 10.01$ GPa by sweeping a magnetic field from 0 to 16 T at ten different temperatures from 100 mK to 8 K (Fig. 7). The MR signal was positive, which ruled out weak localization as the cause of the low-temperature upturn in $\rho(T)$ as had been suggested in the molybdates.$^{20}$ It probably also rules out other mechanisms involving scattering from spins,$^{25}$ and indeed, it may constrain the possible magnetic order on the Ir sublattice. According to Ref. 2, if the Ir moments are ferromagnetically aligned, the ground state is metallic; FM metals usually have a negative MR, however,$^{28}$ in contrast to our observations of a positive signal. So, our results suggest that the ground state is either antiferromagnetic or has the so-called “all-in/all-out” configuration of spins on each Ir tetrahedron. According to Refs. 2 and 3, these magnetic configurations could have a Weyl semimetallic phase separating the Mott-insulating and the metallic ground states.

The weak positive MR signal grows quadratically at low fields ($M \propto H^2$), tends toward saturation at high fields, and becomes smaller with increasing temperature. Such behavior is generic to (nonferromagnetic) metals with closed orbits on the Fermi surface.$^{28}$ The weakness of the MR signal (Fig. 7) may be due to strong scattering, otherwise, a state with small pockets should have large MR due to the small Fermi volume and the low density of carriers. The origin of such strong scattering is not clear, although it is consistent with the fairly large resistivity of these samples, even in the metallic regime; moreover, the metallic state must be unconventional due to the nonmetallic slope of the resistivity $\partial \rho / \partial T < 0$ and the non-Fermi-liquid power-law $T$ dependence.

Further measurements are urgently required in order to elucidate the nature of the negative $\partial \rho / \partial T$ metallic phase. For example, Ref. 2 predicts that the all in/out spin configuration on the Ir tetrahedra has a 1.3-meV gap in the collinear ferromagnetic state. With this gap, our maximum field of 16 T could be sufficient to cause a reorientation of the spins to ferromagnetic, which would cause a huge jump in magnetoresistance, which we do not observe. However, depending on the accuracy of that calculation, a higher magnetic field may be required, so higher field measurements would be informative.

V. CONCLUSION

Using resistivity, we have mapped the temperature-pressure phase diagram of Eu$_2$Ir$_2$O$_7$ between 2 and 12 GPa. The metal-insulator boundary is near 6 GPa. At high temperatures ($T > 100$ K), the resistivity falls by a factor of more than 60 between 2 and 12 GPa, and the behavior crosses over near 6 GPa from an incoherent metal with a very high resistivity and a negative $\partial \rho / \partial T$ to a more conventional metal having a positive $\partial \rho / \partial T$. At intermediate pressures, the crossover is observed as a function of temperature: The sign of $\partial \rho / \partial T$...
changed from positive to negative at a temperature $T^*$ that was not observed using chemical pressure.

The low-temperature behavior evolves from a low-pressure insulating state, having a temperature-dependent gap but a resistivity that does not diverge as $T \to 0$ K to an anomalous metallic state again having a negative $\partial \rho / \partial T$. Conventional explanations for this rising resistivity as $T \to 0$ K, such as the Kondo effect or weak localization, are ruled out. The transition between these ground states is continuous with the critical pressure near 6 GPa, however, there is no temperature scale apparent in the resistivity that vanishes at the critical pressure, rather the high-to-low-temperature crossover persists across the entire phase diagram. An obvious scenario is that the low-temperature phases occur below a magnetic phase transition that is essentially unaffected by pressure.

The high absolute value and the low-temperature rise of the resistivity in the negative $\partial \rho / \partial T$ metallic phase of Eu$_2$Ir$_2$O$_7$ might be manifestations of a topological semimetallic phase.

The anomalous nature of the ground states on both sides of the QCP suggests a topological character for the quantum phase transition and may explain its unusual form. These results should encourage further experiments to test for the existence of topological states near the metal-insulator boundary of the pyrochlore iridates.

ACKNOWLEDGMENTS

The authors acknowledge helpful discussions with Y. B. Kim, Y. Machida, W. Witczak-Krempa, A. Ramirez, and V. Dobrosavljevic. F.F.T. is grateful to Mark Aoshima for his invaluable machining skills. This research was supported by the Natural Science and Engineering Research Council of Canada, the Canadian Institute for Advanced Research, a Grant-in-Aid (Grant No. 21684019) from JSPS, and a Grant-in-Aid for Scientific Research on Priority Areas (Grant No. 19052003) from MEXT, Japan.