Charge density wave and Weyl Semimetal phase in Y$_2$Ir$_2$O$_7$

Abhishek Juyal,1,2,* Vinod Kumar Dwivedi,3,4,* Sonu Verma,1 Shibabrata Nandi,1,5,6 Amit Agarwal,1,† and Soumik Mukhopadhyay1,‡

1Department of Physics, Indian Institute of Technology Kanpur, Kanpur 208016, India
2Georgia Tech Lorraine, IRL 2958-CNRS, 57070 Metz, France
3Materials Science Program, Indian Institute of Technology Kanpur, Kanpur 208016, India
4Department of Physics, Indian Institute of Science, Bengaluru 560012, India
5Forschungszentrum Jülich GmbH, Jülich Centre for Neutron Science (JCN-2) and Peter Grünberg Institut (PGI-4), JARA-FIT, 52425 Jülich, Germany
6RWTH Aachen, Lehrstuhl für Experimentalphysik IVc, Jülich-Aachen Research Alliance (JARA-FIT), 52074 Aachen, Germany

The subtle interplay of band topology and symmetry broken phase, induced by electron correlations, has immense contemporary relevance and potentially offers novel physical insights. Here, we demonstrate charge density wave (CDW) in bulk Y$_2$Ir$_2$O$_7$ for $T < 10$ K, and its transition to the Weyl semimetal (WSM) phase at higher temperatures. The CDW phase is evidenced by a) current induced nonlinear conductivity with negative differential resistance at low temperature, b) low frequency Debye like dielectric relaxation at low temperature with a large dielectric constant $\sim 10^4$, and c) an anomaly in the temperature dependence of the thermal expansion coefficient. The WSM phase at higher temperature is confirmed by the DC and AC transport measurements which show an inductive response at low frequencies. More interestingly, we show that by reducing the crystallite size, the low temperature CDW phase can be eliminated leading to the restoration of the WSM phase.

Strong electron-electron correlation and topology are generally considered mutually exclusive domains of physics. However, recent observations of interaction driven topological phase transition [1, 2] have opened up prospects for exploring the interplay of correlation and topology. In fact, Weyl semimetals (WSM) were first predicted in correlated pyrochlore iridates, R$_2$Ir$_2$O$_7$ ($R = Y, Eu, Nd$) [3]. These 5$d$ orbital based iridates can host a variety of topological and quantum phases in addition to the WSM phase [3–13]. The search for Fermi arc surface states in Y$_2$Ir$_2$O$_7$ and other iridates has been unsuccessful due to the unavailability of good quality single crystals and inadequate surface preparation rendering techniques such as ARPES and STM, ineffective. However, there could be a more fundamental reason: Weyl nodes could gap out forming density wave instabilities due to chiral symmetry breaking induced by coulomb interaction [14–16]. Recently, a large negative contribution to the longitudinal magnetoresistance in the sliding mode of the charge density wave (CDW) phase of WSM candidates including nanorods based on Y$_2$Ir$_2$O$_7$ have been observed [15, 17]. The negative longitudinal magnetoresistance in such cases originates from the axionic contribution of the chiral anomaly to the phason current [15, 17].

Apart from Y$_2$Ir$_2$O$_7$, amongst the iridates family, Nd$_2$Ir$_2$O$_7$ shows a quadratic band touching at the $\Gamma$ point which gaps out at low temperature [18]. The claim of metal-semimetal transition in the optical conductivity data for Eu$_2$Ir$_2$O$_7$ [19] needs to be backed by other more compelling evidence. Epitaxial strain induced ‘all-in, all-out’ ordering in thin films of Pr$_2$Ir$_2$O$_7$ breaks the time reversal symmetry, possibly leading to WSM phase [20]. Although single crystalline nanorods of Y$_2$Ir$_2$O$_7$ have shown evidence of possible chiral anomaly in the gapped out WSM phase [14, 15], the question remains whether the conclusions drawn can be extended to bulk Y$_2$Ir$_2$O$_7$ as well.

Here, we present experimental evidence of gapped out WSM ground state in bulk Y$_2$Ir$_2$O$_7$, showing characteristics of (axionic) CDW with a distinct transition to WSM phase above 10 K. Both the CDW phase in bulk Y$_2$Ir$_2$O$_7$ for $T < 10$ K, and the WSM phase above $T > 10$ K are confirmed independently via DC and AC transport measurements. The WSM to CDW transition is also observed in the thermal expansion experiments. The CDW gap opening can be prevented by reducing the grain size, thus extending the WSM state to low (~1 K) temperatures. Remarkably, we find the DC transport in nano-crystalline samples to be governed by the Coulomb interaction induced diffusive transport in the WSM phase [21].

Electrical transport measurements were performed on bulk...
and nano-crystalline $Y_2Ir_2O_7$ samples. See supplementary material (SM) [22] for details of sample preparation and characterization. To start with, we present the evidence for the CDW phase in the bulk sample, that is observed in current-driven nonlinear IV characteristics shown in Fig. 1. The linear conductivity at low voltage is followed by the onset of nonlinear conduction at higher voltages when the CDW starts sliding. More interestingly, the non-ohmic IV characteristics at 1.3 K also exhibits current controlled negative differential resistance (NDR). Both of these effects are associated with CDW de-pinning, and the resultant constant current IV characteristic showing NDR is commonly referred to as ‘S-shaped’ response. The NDR is not observed at higher temperatures although the nonlinear IV characteristic is found to persist up to 9 K in Fig. 1(a).

The essential features of these measurements are captured by a simple model describing the dynamics of the CDW phase. The CDW phase may be weakly pinned by impurities, lattice defects or confined by the grain boundaries [23]. Electrical transport occurs due to the translational motion of the CDW and that of the normal carriers. At low electric field (or current) values below a critical threshold, $E < E_T$ (or $J < J_T$), normal quasiparticles carry the current leading to ohmic behaviour while the CDW domains remain pinned. For $E > E_T$ (or $J > J_T$), the CDW domains become partially de-pinned, leading to nonlinear contribution to conductivity. In this regime, the transport is described by Bardeen’s CDW tunneling theory [24]. The threshold field directly probes the effectiveness of the pinning of the CDW order parameter. We find $E_T(T) = E_0 \exp(-\Delta/T)$, with $T_0 \approx 3$ K denoting the strength of the pinning potential [25], as shown in the inset of Fig. 1(a).

On increasing the current (or field) beyond a second threshold, $J > J_T$ (or $E > E^*_T$), the CDW dislodges and moves with a single phase. Consequently, for $J > J_T$ the voltage across the sample drops abruptly as the current increases. This leads to NDR as shown in Fig. 1(a) for $T = 1.3$ K. In this regime, the conductivity is dominated by coherent CDW transport, which is well described by the damped dynamics of the CDW phase [26, 27]. In a constant current experiment, the nonlinear I-V characteristics above the de-pinning threshold $E^*_T$ is given by [26, 28]

$$E = \frac{J}{\sigma_n} - \frac{\beta E^*_{\Delta}}{1 + \beta} \left( \frac{J}{J^*_{\Delta}} \right)^2 - \frac{1}{2}.$$  

Here $\sigma_n$ is the ohmic conductivity of the normal quasiparticles, and $\beta$ is a system and temperature dependent parameter. The conductivity $dE/dJ$ has a discontinuity at the NDR threshold $J^*_T$. This discontinuity is explicitly shown for the $T = 1.3$ K curve in both panels of Fig. 1 (b), along with the corresponding fit to Bardeen’s tunneling theory for $J_T < J < J^*_T$ and to Eq. (1) for $J > J^*_T$. This is possibly the first demonstration of the coexistence of quantum and classical regime of the CDW transport. Interestingly, the IV characteristics in the NDR regime also show negative (positive) longitudinal (transverse) magneto-resistance, similar to that found in single crystalline nanorods of $Y_2Ir_2O_7$ [15]. This is shown explicitly in Fig. S3 in SM [22].

We measure the low-frequency dielectric response using the standard lock-in technique. For $T < 10$ K, the AC transport measurements show distinctly different behaviour compared to the measurements at higher temperatures, as highlighted in Fig. 2. In particular, we find that for $T < 10$ K, the dielectric constant shows a Debye like relaxation indicating a CDW phase. Similar to the observations in single crystalline $Y_2Ir_2O_7$ nanorods [14, 15], we find that the DC conductivity and the peak frequency ($\omega_p$) of the imaginary part of the dielectric constant show an Arrhenius-like temperature dependence: $\sigma_0 \propto \omega_p \propto \exp(-\Delta/T)$, with $\Delta = 16.1$ K. Physically this occurs due to the screening of the damped collective charge oscillations of the CDW by the thermally excited normal carriers [29, 30]. This gives us a mean-field estimation of the CDW transition temperature (2$\Delta = 3.52 \times T_{CDW}$) to be $T_{CDW} \approx 9.15$ K, consistent with the DC measurements. The corresponding coherence length can be estimated to be $\xi_0 \approx 2.4$ nm, which is much smaller than the grain size~$\approx 1 \mu$m, and similar to the observed value in other polycrystalline samples [31].

For $T > 10$ K, the real component of the conductivity [see Fig. 2(a)] displays behaviour similar to that of the Weyl...
The CDW to WSM transition in Y$_2$Ir$_2$O$_7$ can also be seen from the temperature dependence of the DC conductivity. To distinguish the nature of DC transport between activated and power law behaviour, we plot $-\rho^{-1} \frac{dT}{d\rho}$ as a function of T. The activated behaviour of the DC transport for $T < 10$ K (with a slope $\approx -2$ in the CDW phase) and a power law behaviour above $T > 10$ K (with a slope $\approx -1$ in the WSM phase) is clearly established. The CDW transition temperature estimated by fitting the activated DC transport [$\rho \propto e^{\Delta T_{CDW}/(k_B T)}$] below $T < 10$ K is consistent with the earlier estimated value.

For $T > 10$ K, a clear power law behaviour in the resistivity, $\rho(T) \propto T^{-n}$, is found till $T \approx 80$ K. However, different experiments have reported varying power-laws for the resistivity of Y$_2$Ir$_2$O$_7$. While our bulk samples show $n = 3.02$, close to the value of $n = 3.1$ reported in [32], there have also been reports of $n = 4.3$ [33], $n = 4.6$ [34] and $n = 4.8$ [35]. See Fig. 3(b) for a comparison, where we estimate the exponents by digitizing and fitting all the available data-sets up to 50 K, for uniformity. This clearly shows that the low temperature power-law behaviour of $\rho(T)$ in Y$_2$Ir$_2$O$_7$ is non-universal, and possibly dependent on various factors such as crystallite size, disorder layout, etc.

The nano-crystalline samples show a power law exponent close to unity, which has also been predicted to arise in the Coulomb interaction dominated diffusive transport regime of WSM. The predicted DC conductivity [21] is given by

$$\sigma_{dc}(T) = \frac{e^2}{\hbar} \frac{k_B T}{h v_F(T)} \frac{0.45}{\alpha T^{\alpha}}.$$

Here, the renormalized fine structure constant is $\alpha = \alpha_1 [1 + (N + 2)\alpha_1 (L \Lambda^{-1})]$ and the renormalized Fermi velocity is $v_F(T) = v_F (\alpha_1 / \alpha T)^{2+2/N}$. The number of Weyl nodes is specified by $N = 24$, and $\Lambda$ is a momentum cutoff set by the separation between the Weyl nodes. This model description for $\rho(T)$ [21] fits our experimental data for the nano-crystalline sample, remarkably well over the entire temperature range, as shown in Fig. 3(c).
FIG. 4: a) Variation of the relative length ($\Delta \tilde{L} = \Delta L/\Delta L_{T=180}$) and $\alpha (\tilde{\alpha} = \alpha/\alpha_{T=180})$ with $T$ for the bulk polycrystalline $Y_2Ir_2O_7$. The thermal expansion coefficient (blue curve) fits well with the Debye model (red curve) at high $T$. However, it shows an anomaly at low temperature coinciding with the $T_{\text{CDW}}$ obtained from the transport measurements. This is highlighted in the zoomed panel b) showing the low temperature regime. Here, we have shifted the $\Delta L$ curve and the different linear fits below and above $T_{\text{CDW}}$ are shown by red lines. c) The deviation of $\alpha/T^3$ from the Debye behaviour (dashed black line) at low $T$.

This naturally prompts the question: why does the CDW gap close on reducing the particle size (or increasing the surface to volume ratio), leading to the restoration of the WSM phase in $Y_2Ir_2O_7$ [36]? The possibility of the quantum confinement gap [$\Delta_{QC} \approx h^2/(m_e L^2)$] becoming larger than the CDW gap is unlikely in this case as, even for an average particle size of 50 nm we have $\Delta_{QC} \sim 0.03$ meV, which is almost 30 times smaller than $k_BT_{\text{CDW}} \approx 0.86$ meV. We rule out the possibility of the softening of phonon mode due to increased surface contribution by our specific heat measurements (not shown here), which shows an enhancement in the $\theta_D$ in the nano-crystalline sample ($\theta_D = 503.9 \pm 14.5$ K) compared to the corresponding bulk value ($\theta_D = 402.6 \pm 9.1$ K). One possibility supported by our X-ray spectroscopy data is the increase in carrier kinetic energy in nano-crystalline samples owing to the enhancement of the concentration of higher oxidation states - $I^{5+}$ (See Fig. S2 in SM) or due to quantum confinement effects. The increased kinetic energy in nano-crystalline samples is likely to reduce the impact of correlation effects and reduce the $T_{\text{CDW}}$. Recently pressure induced closing of a large CDW gap has been observed in Ta$_2$Se$_8$I, which exhibits a Weyl semimetal to CDW transition close to room temperature in ambient pressure [37]. In our case, the CDW gap being much smaller, increased surface pressure due to reduction of particle size could be effectively playing a similar role.

For independent confirmation of the CDW phase transition in $Y_2Ir_2O_7$, we performed thermal expansion measurements using a capacitive dilatometer having sub-angstrom resolution with high sensitivity of $\Delta L/L \sim 10^{-10}$ at low temperature. We measured the linear thermal expansion coefficient, $\alpha(T) \equiv dL/dT$, and verified it for different heating and cooling rate. The relative thermal expansion $\Delta L/L$, shown in Fig. 4, decreases monotonically with the lowering of temperature. However, there is an anomaly at $T \approx 10.4$ K (consistent with the estimated $T_{\text{CDW}} \approx 9.15$ K from transport measurements) with relative dilatation of about $1.02 \times 10^{-6}$, as shown in Fig. 4 (a)-(b). The anomaly is highlighted further in the thermal expansion coefficient which shows a sharp minimum at the same temperature. The $\alpha(T)$ curve fits reasonably well to the Debye model at higher temperature (see SM [22] for details). The deviation from the Debye $T^3$ law in $\alpha(T)$ is highlighted by plotting the residual $\alpha(T)/T^3$ vs $T$ on the log-log scale in Fig. 4 (c). Similar deviation from the $T^3$ law has also been observed in low temperature specific heat for other CDW systems [38–40]. This deviation can be attributed to the splitting of the acoustic phonon mode (for $T < T_T$) into the gapped amplitude mode [which gives the ‘hump’ for $T \sim T_T$ in $\alpha(T)/T^3$ plot in Fig. 4(c)] and the gapless phase mode ($T \rightarrow 0$ behaviour of the $\alpha(T)/T^3$ curve) of the CDW condensate.

To summarize, we present clear experimental evidence of temperature and grain-size dependent phase transition from the CDW phase to the WSM phase in bulk $Y_2Ir_2O_7$ using DC and AC transport experiments. The phase transition is independently confirmed via thermal expansion measurements as well. The low frequency dielectric response in the WSM phase at high temperature is consistent with the theoretically predicted response in WSM having short range neutral disorder. More interestingly, we demonstrate that the CDW gap is suppressed significantly on reducing crystallite size. The observed DC conductivity over a wide temperature range in the nano-crystalline $Y_2Ir_2O_7$ shows interaction induced diffusive behaviour characteristic of WSM. The grain size dependent tunability of the phase transition clearly suggests the existence of a quantum critical point separating the WSM phase and the broken symmetry (possibly axionic) CDW phase. This opens up new directions for exploring quantum criticality and interaction driven interplay between topological and symmetry broken phases.

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* These authors contributed equally

1 Electronic address: amitag@iitk.ac.in

2 Electronic address: soumikm@iitk.ac.in

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