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Infrared evidence of a Slater metal-insulator transition in NaOsO_3

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The magnetically driven metal-insulator transition (MIT) was predicted by Slater in the fifties. Here a long-range antiferromagnetic (AF) order can open up a gap at the Brillouin electronic band boundary regardless of the Coulomb repulsion magnitude. However, while many low-dimensional organic conductors display evidence for an AF driven MIT, in three-dimensional (3D) systems the Slater MIT still remains elusive. We employ terahertz and infrared spectroscopy to investigate the MIT in the NaOsO_3 3D antiferromagnet. From the optical conductivity analysis we find evidence for a continuous opening of the energy gap, whose temperature dependence can be well described in terms of a second order phase transition. The comparison between the experimental Drude spectral weight and the one calculated through Local Density Approximation (LDA) shows that electronic correlations play a limited role in the MIT. All the experimental evidence demonstrates that NaOsO_3 is the first known 3D Slater insulator.

In the last sixty years considerable effort has been devoted to the study of the MIT in transition metal oxides^{1,2}. In their fundamental papers Mott³ and Hubbard⁴ proposed that the electron-electron interaction (U) is the key ingredient of the MIT. Electronic correlations may strongly renormalize the kinetic energy (t) of charge-carriers leading to the formation of a gap at the Fermi energy and then to an insulating state regardless of magnetic correlations. Temperature and/or pressure may increase the t/U ratio, thus inducing a transition (Mott-Hubbard transition) from the insulating to the metallic state. V_2O_3 ⁵, $\text{Ni}(\text{S},\text{Se})_2$ ⁶ and NdNiO_3 ⁷ are considered textbook examples of Mott-Hubbard materials. On the other hand, Slater⁸ suggested a different mechanism for the MIT: he stated that antiferromagnetic order alone could open up a gap. The establishment of a commensurate AF order induces an opposite potential on each nearest electron site which doubles the magnetic unit cell and cuts the first Brillouin zone. This results in a splitting of the occupied bands and, in case of half-filling, in a gap for charge excitations. So far the only evidence of magnetically driven MITs was found on low dimensional organic compounds as Bechgaard salts⁹⁻¹¹, while the 3D candidates $\text{Cd}_2\text{Os}_2\text{O}_7$ ¹²⁻¹⁴ and $\text{Ln}_2\text{Ir}_2\text{O}_7$ ¹⁵ could not be classified as Slater insulators due to magnetic frustration in their pyrochlore lattice¹⁶. Experimental indications of a Slater transition have never come up until last year¹⁷. By using neutron and x-ray scattering, Calder *et al.* have shown that in the perovskite NaOsO_3 the MIT at $T_{\text{MIT}} = 410$ K, previously discovered on the basis of transport and magnetic measurements¹⁸, is concomitant with the onset of long-range commensurate three-dimensional antiferromagnetic order. As a perovskite with octahedral environment Os^{5+}O_6 , NaOsO_3 shows a half-filled $5d^3$ electronic configuration. In addition, $5d$ orbitals are far more spatially extended than those in $3d$ systems, thus electron-electron interactions is expected to play a minor role in this compound, as also recently argued from theoretical calculations^{19,20}. In this work we probe the low-energy electrodynamics of NaOsO_3 across the MIT by terahertz and infrared spectroscopy. From an analysis of the optical spectral weight we establish that NaOsO_3 is a weakly correlated material²¹. We also show that, at variance with a Mott-Hubbard MIT^{5-7,22}, the optical conductivity does not vanish at T_{MIT} as the charge gap opens up in a continuous way in agreement with the second order character of the MIT. Those experimental results definitively clarify the nature of the three-dimensional MIT in NaOsO_3 in terms of the Slater mechanism.

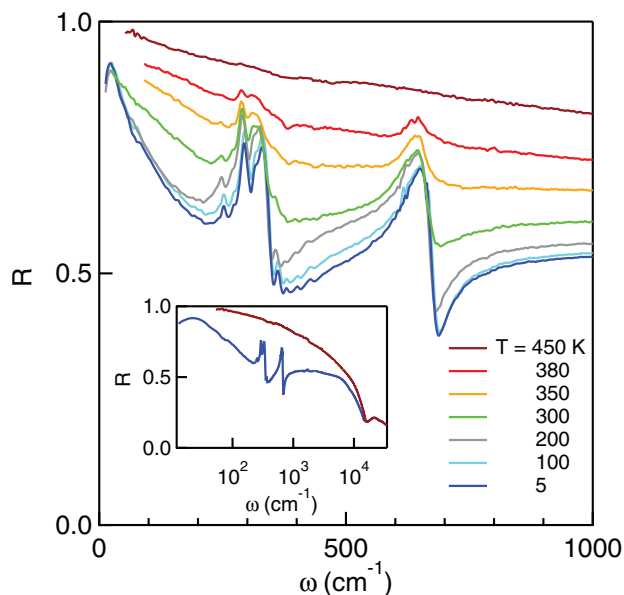


Figure 1 | The near-normal incidence reflectance of NaOsO₃ is reported at selected temperatures from 5 K to 450 K on a linear scale in the 0–1000 cm⁻¹ frequency range. In the inset data are shown in the insulating (5 K) and in the metallic (450 K) states on a log scale in the whole frequency range. The MIT temperature is 410 K.

Results

The reflectance $R(\omega)$ of NaOsO₃ is shown in Fig. 1 in the 0–1000 cm⁻¹ frequency range. At 450 K it shows a metallic response, approaching unity at zero frequency. After crossing T_{MIT} this metallic behavior depletes progressively and, in the antiferromagnetic insulating phase, two complex phononic structures start to appear as a consequence of the reduced screening. At 5 K there is a well visible double phonon peak with characteristic frequencies around 300 cm⁻¹ and 330 cm⁻¹ and another phonon resonance at 650 cm⁻¹. Moreover, $R(\omega)$ shows a rising tail for $\omega \rightarrow 0$ related to a low-frequency mode centered around 20 cm⁻¹ (see discussion below). In the inset of Fig. 1 we show instead the reflectance over the entire measured range in the metallic (450 K) and in the insulating state (5 K). It is well evident that the MIT determines a strong modification of the electronic properties of NaOsO₃ over a frequency scale up to nearly 10000 cm⁻¹.

The main panel of Fig. 2 shows the optical conductivity $\sigma_1(\omega)$ on a linear scale in the 0–10000 cm⁻¹ frequency range as obtained from reflectance data by Kramers-Kronig relations. The same quantity is represented in the inset on a logarithmic scale for selected temperatures above and below T_{MIT} . The symbols on the vertical axis indicate the dc conductivity values calculated from resistivity data measured on samples coming from the same batch¹⁸. At all temperatures there is a good agreement between the zero frequency limit of $\sigma_1(\omega)$ and the measured σ_{dc} .

The optical conductivity at 450 K shows a metallic behavior with a broad pseudo plasma-edge around 12000 cm⁻¹ which separates the low energy excitations from a huge interband transition around 20000 cm⁻¹. This absorption band is mainly associated to charge-transfer excitations among Os 5d and O 2p states^{18,19}. Below T_{MIT} the metallic conductivity sharply decreases in the far-infrared through a transfer of spectral weight (SW) to a mid-infrared (MIR) band centered around 3000 cm⁻¹. The low-frequency SW depletion is nearly exhausted at 200 K where the MIR is located at about 4000 cm⁻¹.

A broad feature centered around 22 cm⁻¹ can be seen in the insulating phase between 5 and 200 K and it is probably hidden at higher-T by the free carrier background. We associate this peak to an antiferromagnetic resonance. Indeed, the insulating phase of NaOsO₃ corresponds to a G-type AF configuration¹⁷ where the spins are

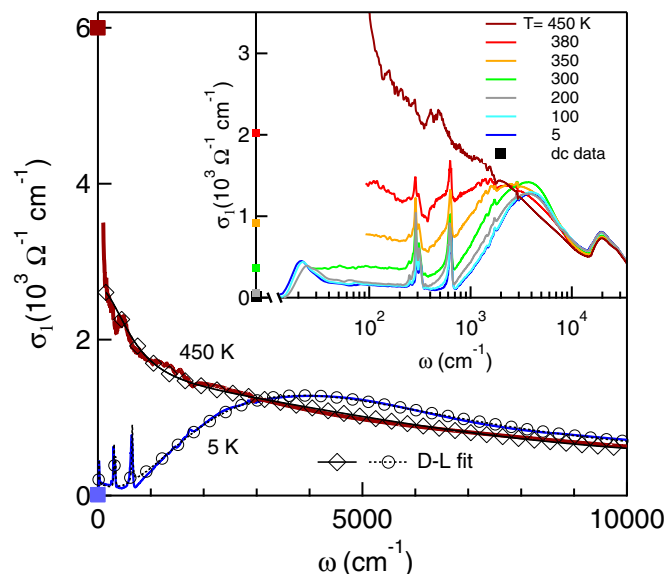


Figure 2 | Optical conductivity of NaOsO₃ at 5 K and 450 K on a linear frequency scale as obtained from Kramers-Kronig transformations. Symbols on the vertical axis stand for dc values of the conductivity, calculated from transport measurements. The inset shows the optical conductivity on a log scale at selected temperatures. The Drude-Lorentz fitting results (see text) are indicated by open symbols.

oriented along the c -axis. The antiferromagnetic mode corresponds to a precession of spins along c induced by some degree of magnetic anisotropy^{23,24}.

The loss of SW in the far-infrared mirrors the opening of an optical gap E_g already distinguishable at 380 K, whose size increases for decreasing T . One can then extract E_g from data in Fig. 2 and compare its temperature dependence with that expected for a second order (Slater) phase transition. The optical conductivity in the insulating AF phase can be described through the following equations¹³:

$$\sigma_1(\omega) = m\omega + q \quad \text{for } \omega \leq E_g \quad (1)$$

and

$$\sigma_1(\omega) = A[\omega - E_g(T)]^\alpha \quad \text{for } \omega \geq E_g \quad (2)$$

Here, the linear term describes the low-frequency background which is mainly due to thermal excited charge-carriers across the insulating gap (see below). For higher frequencies instead, we assume a behavior of $\sigma_1(\omega)$ similar to that of a semiconductor in the presence of direct band to band transitions²⁵. The curves thus obtained, reported as dashed lines in the inset of Fig. 3, nicely fit the rising edge of $\sigma_1(\omega)$ at all temperatures with $\alpha = \frac{1}{2}$. An estimate of the gap $E_g(T)$ is then obtained through the intersection between Eq. 1 and Eq. 2 and its temperature behavior, which is plotted as empty circles in Fig. 3, resembles that of a Bardeen-Cooper-Schrieffer (BCS) function in good agreement with the second order character of the MIT. Let us mention that similar gap values can be also achieved by substituting the linear conductivity (Eq. 1) with a Drude term. Therefore the error bars in Fig. 3 take into account the small variation of $E_g(T)$ in terms of different extracting methods and the smearing effect in the conductivity edge due to the temperature.

A BCS-like analytic expression for $E_g(T)$ can be written as²⁵:

$$\frac{E_g(T)}{E_g(0)} = \tanh \frac{E_g(T)T_{MIT}}{E_g(0)T} \quad (3)$$

This function well describes data in Fig. 3 furnishing a value of $E_g(0) = 825 \pm 25$ cm⁻¹ and a $T_{MIT} = 400 \pm 10$ K in fair agreement with

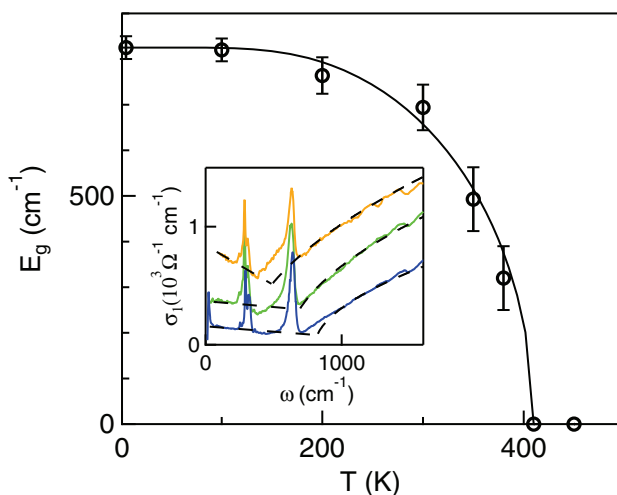


Figure 3 | Magnitude of the optical gap with its fit to Eq.(3). The extrapolations of the optical conductivity based on Eq. (1) and (2) are shown in the inset as dashed curves at $T = 350, 300, 5$ K (from top to bottom).

the value ($T_{MIT} = 410$ K) obtained through transport and magnetic measurements^{17,18}. Moreover, $E_g(0)/k_B T_{MIT} = 3.0 \pm 0.1$, thus suggesting a weak coupling regime in agreement with a Slater scenario.

As mentioned before, the gap opening corresponds to a loss of SW in the infrared which is transferred to higher frequencies. The energy scale involved in this process can be estimated from Fig. 4 where the optical spectral weight is reported. It can be written as follows:

$$SW(\omega_c, T) = \int_0^{\omega_c} \sigma_1(\omega, T) d\omega \quad (4)$$

and it is proportional to the number of carriers taking part to the optical absorption up to a cutoff frequency ω_c . The SW is nearly conserved for $\omega_c \simeq 15000$ cm^{-1} . This energy scale is well captured by the LDA + U calculation reproducing, for a G-type antiferromagnetic order, the insulating state for a moderate value of $U \sim 1-2$ eV^{17,19,20}.

The SW redistribution can be analyzed in detail by fitting $\sigma_1(\omega)$ through a multi-component Drude-Lorentz (D-L) model. The complex optical conductivity is written in terms of a Drude contribution and five Lorentzian curves as:

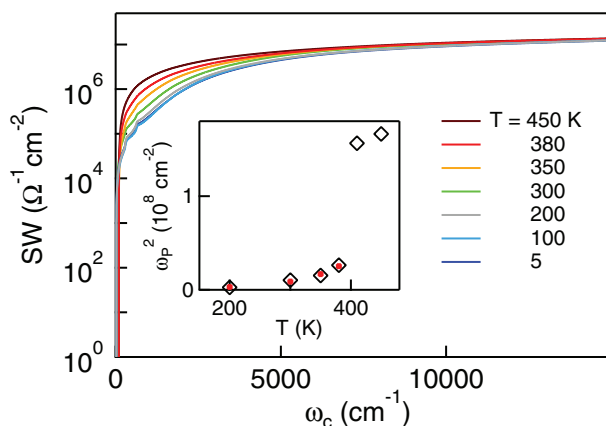


Figure 4 | Optical spectral weight vs cutoff frequency ω_c at selected temperatures. The SW is recovered at nearly 15000 cm^{-1} . Inset: the squared plasma frequency vs T behavior is the result of the Drude-Lorentz fit analysis of the optical conductivity. Values calculated from the relation $Ae^{-E_g(T)/2k_B T}$ are indicated by red symbols.

$$\tilde{\sigma}(\omega) = \frac{\omega_p^2 \tau}{4\pi(1 - i\omega\tau)} + \frac{\omega}{4\pi i} \sum_j \frac{S_j^2}{\omega_j^2 - \omega^2 - i\omega\gamma_j} \quad (5)$$

In the Drude term ω_p is the plasma frequency, τ is the scattering time, while the Lorentz oscillators are peaked at finite frequencies ω_j with strength S_j and width γ_j . We used a Lorentzian oscillator both for the mid-infrared band and for the charge-transfer transition around 20000 cm^{-1} . The phonon absorption is described in terms of three Lorentzian components. Examples of D-L fits are represented in the main panel of Fig. 2 both at 450 K (metallic phase) and 5 K (insulating phase). From the fit we have extracted ω_p^2 whose values vs. T are shown in the inset of Fig. 4 as open diamonds. One can observe a drastic collapse of ω_p^2 at the MIT. Below T_{MIT} the residual Drude weight is mainly determined by the charge-carriers thermally excited above the insulating gap. Indeed ω_p^2 can be fitted by an exponential T-dependence $\omega_p^2(T) = Ae^{-E_g(T)/2k_B T}$ (red symbols) with $A = 4.6 \cdot 10^7$ cm^{-2} , nearly corresponding to the square of the plasma frequency at 410 K. By employing the gap values in Fig. 3, this exponential equation properly reproduces the experimental points from 200 K, where the Drude contribution is still zero, to 380 K where the gap is nearly closed.

Even more importantly the plasma frequency at 450 K can be used for a quantitative determination of the degree of electronic correlation in NaOsO_3 . Indeed the Drude plasma frequency is related to the experimental kinetic energy of mobile carriers by the relation $K_{exp} \sim \omega_p^2/8$. Due to correlation effects this value is renormalized (reduced) with respect to the kinetic energy provided by Local Density Approximation calculations K_{LDA} ²¹. In a Mott-Hubbard insulator $K_{exp} = 0$ as charge-carriers are completely localized due to Coulomb correlation, at variance with the band-structure results predicting a metallic response with a finite K_{LDA} . Conversely, for conventional metals like gold and copper, the ratio $K_{exp}/K_{LDA} \sim 1$, indicating the scarce relevance of electronic correlations. From the Drude-Lorentz fit at 450 K shown in the main panel of Fig. 2 we estimate $\omega_p \sim 1.6$ eV; LDA calculations provide $\omega_p(\text{LDA}) = 2.1$ eV instead. From these data we achieve a ratio $K_{exp}/K_{LDA} \sim 0.6$, thus locating NaOsO_3 at the border between correlated metals and uncorrelated systems suggesting a relatively weak electronic correlation.

Discussion

NaOsO_3 shows a half-filled $5d^3$ electronic configuration. Since $5d$ orbitals are spatially more extended than those in $3d$ and $4d$ systems, theoretical calculations argue that electron-electron interactions should play a minor role in the low-energy electrodynamics suggesting a Slater origin of the MIT^{19,20}. In this paper we provide robust experimental evidence for the Slater mechanism. Indeed, the K_{exp}/K_{LDA} ratio, which has been demonstrated to well estimate the electronic correlation degree in many materials, gets a value $K_{exp}/K_{LDA} \sim 0.6$. This firstly suggests that NaOsO_3 lies near the band-like itinerant regime²¹. Secondly, as demonstrated in Fig. 3, the gap develops in a continuous way in NaOsO_3 , by following a BCS-like temperature dependence. Conversely, in Mott-Hubbard materials as V_2O_3 , $\text{Ni}(\text{S,Se})_2$ and NdNiO_3 , a gap abruptly appears while crossing T_{MIT} ²². Moreover, both the $E_g(0)/k_B T_{MIT} = 3.0 \pm 0.1$ ratio and the gap temperature dependence are in good agreement with the second order character of a Slater MIT. Finally, the concomitance of the MIT with the AF ordering, as measured through neutron scattering, suggests that magnetism is the main cause for the MIT. All these findings clarify the MIT in terms of the Slater mechanism, showing that NaOsO_3 is the first known 3D Slater insulator.

Methods

Sample preparation. A high-density polycrystalline pellet sample of NaOsO_3 was synthesized starting from Na_2O_2 and OsO_2 powders through a high pressure



technique (see Ref. 18). A powder x-ray diffraction study using Cu $K\alpha$ radiation confirmed the absence of impurities.

Optical measurement. We performed near-normal reflectance measurements on an accurately polished sample using a Michelson interferometer in the frequency range from 10 cm^{-1} (this limit varies for some temperatures) to 15000 cm^{-1} . Terahertz and subterahertz ($\omega < 50\text{ cm}^{-1}$) measurements were performed using radiation produced at the SISSI²⁶ and IRIS²⁷ infrared beamlines at Elettra and Bessy-II synchrotrons respectively. A gold or silver (depending on the spectral range) coating was evaporated *in situ* over the sample surface and used as a reference. The optical conductivity was calculated by Kramers-Kronig transformations. Low-frequency reflectance data were extrapolated with standard methods (Hagen-Rubens or constant lines) taking into account the resistivity dc values measured in samples of the same batch. A high-frequency tail¹³ was instead merged to the data above 15000 cm^{-1} .

LDA calculation. The K_{LDA} was obtained from first-principles calculations within Local Density Approximation (LDA) of density functional theory. The electronic structure was calculated from full-potential augmented plane wave (APW) methods with the WIEN2k package²⁸. The spin-orbit interaction was included within self-consistent calculations. The lattice constants and atomic positions were taken from experimental values¹⁸. The muffin-tin radii were chosen as 2.0, 1.8 and 1.7 a.u. for Na, Os, and O, respectively. The cutoff wave number K in interstitial region was set to $RK = 8$, where R is the smallest atomic radius, i. e., 1.7 a.u. for O. The integration over Brillouin zone was performed by a tetrahedron method with up to 1960 k -points in an irreducible zone. The optical conductivity and plasma frequency were estimated from the momentum matrix elements as described in Ref. 29. Since NaOsO_3 has orthorhombic symmetry with $Pnma$ space group, the optical conductivity tensors are characterized by three diagonal elements σ_{xx} , σ_{yy} , and σ_{zz} . Accordingly, we obtained plasma frequency for each direction as $\omega_{p_{xx}} = 2.2\text{ eV}$, $\omega_{p_{yy}} = 1.8\text{ eV}$, $\omega_{p_{zz}} = 2.4\text{ eV}$. The averaged plasma frequency $\omega_p(\text{LDA}) = (\omega_{p_{xx}} + \omega_{p_{yy}} + \omega_{p_{zz}})/3$ was estimated as 2.1 eV.

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Author contributions

Y.S. and K.Y. fabricated and characterized NaOsO_3 samples. I.L.V., A.P., P.D.P., O.L., U.S. and S.L. carried out the terahertz experiments and data analysis. M.A. was responsible of the LDA calculations. S.L. was responsible for the planning and the management of the project with inputs from all the co-authors, especially from I.L.V., A.P. and K.Y. All authors extensively discussed the results and the manuscript that was written by I.L.V., A.P. and S.L.

Additional information

Competing financial interests: The authors declare no competing financial interests.

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