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*Ab initio* calculations and ellipsometry measurements of the optical properties of the layered semiconductor $\text{In}_4\text{Se}_3$

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Authors
In this work, we present a thorough study of the optical properties of the layered orthorhombic compound In$_4$Se$_3$. The dielectric function—real and imaginary parts, the complex refraction index, the reflectivity, the absorption coefficient, and the conductivity of In$_4$Se$_3$ were calculated with the inclusion of the spin-orbit interaction, using an *ab initio* FP-LAPW method based on DFT. Also, generalized ellipsometry was employed for more precise measurement of the anisotropic dielectric functions for polarization along crystal $a$, $b$, and $c$ axes of orthorhombic absorbing In$_4$Se$_3$ single crystals cut approximately parallel to (100) at photon energies from 0.76 to 3.1 eV. Our experimental results show a good agreement with our calculations. We discuss the location and nature of the main optical peaks appearing in the spectra. The obtained optical functions display a rather anisotropic behavior, mainly in the infrared–visible region. Our results seem to be predictive to a high extension, given the scarce experimental information about its optical properties.

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I. INTRODUCTION

The In$_4$Se$_3$ orthorhombic crystal ($D_{12}^{12}$) is a narrow band gap semiconductor containing 27 atoms in the unit cell (see Fig. 1) and much is known about its band structure. It crystallizes in a layered structure with, in principle, weak interaction of the van der Waals type between the layers and strong covalent–ionic interactions within the layers.1

The material seems to have been less studied among the layered chalcogenides crystals, yet continues to attract attention as a natural low dimensional quasi-two-dimensional (quasi-2D) material in much the same way as occurs with layered systems with flat layers held together with van der Waals forces. Indeed, it was suggested that it could be suitable for fabrication of infrared optical fiber and nanowires, and a recent article reports an In$_4$Se$_3$-related material (In$_4$Se$_3$ self-doped with Se deficiencies) with a high thermoelectric performance as an important candidate for recycling waste heat into useful electricity. In addition, In$_4$Se$_3$ polycrystalline thin films have been synthesized and suggested for photovoltaic applications.7,8

Also, other experimental [e.g., Auger spectroscopy,9 and single crystal surface characterization with STM, LEED, and XPS (Ref. 3)] and theoretical (e.g., lattice dynamics modelization,10 pseudopotential band structure calculations11) studies have been reported. In previous work,12–14 we studied both the surface and the bulk electronic states, showing that the system does have a bulk band structure (i.e., discernable and significant band dispersion) perpendicular to the cleavage plane, with an anisotropic dispersion of about 1 eV in the In chain direction.12 In spite of all these known details of the band structure, to the best of our knowledge neither experimental data (with the exception of far-infrared optical properties15–17) nor theoretical calculations of wide range optical spectra of the In$_4$Se$_3$ are available in the literature. Therefore, and since state-of-the-art calculations have sensibly improved, an exhaustive and updated theoretical study of the In$_4$Se$_3$ optical properties based on *ab initio* calculations is quite desirable.

In this work, we present a thorough study of the optical properties of the system: the dielectric function—real and imaginary parts, the complex refraction index, the reflectivity, the absorption coefficient, and the conductivity of In$_4$Se$_3$ were calculated with the inclusion of the spin-orbit interaction. Based on our previous electronic structure results,12,14 we discuss the location and nature of the main optical peaks appearing in the spectra. As a companion to the theory, we report the determination of the intrinsic orthorhombic optical properties for photon energies from 0.76 to 3.1 eV of In$_4$Se$_3$ single crystals cut approximately parallel to (100). Biaxial optical properties of anisotropic bulk and thin film materials can be obtained by generalized ellipsometry. This concept has been demonstrated recently for numerous materials, and particularly for the orthorhombic,18–20 monoclinic,21,22 and triclinic23 materials. Our experimental results show a good agreement with our calculations.

We obtained optical properties with rather anisotropic behavior, mainly in the infrared–visible region, which can be explained with our model. Our results predict the optical properties on this system, to much higher energies than currently available from experiment.
We modeled the In₄Se₃ in the framework of the density functional theory (DFT). In this work, we used the WIEN2K package, which self-consistently finds the eigenvalues and eigenfunctions of the Kohn-Sham equations for the system. As it is well known the local density approximation (LDA) (e.g., in the Perdew-Wang parametrization) for the exchange and correlation potential has had great success in dealing with the calculation of electronic properties. However, it does not accurately describe some important properties, for example, there is a tendency to underestimate the band gaps. A formal correction is obtained by including the gradient of the charge density. This is the so-called generalized gradient approximation (GGA), which we used in the formal Engel-Vosko scheme. We also tested the Perdew-Burke-Ernzerhof (PBE) parametrization scheme but found, similarly to other authors, that the Engel–Vosko scheme gives improved results of the band gap, so we present here only these latter. However, we present in Table I our band gaps both with PBE and EV-GGA scheme, along with other reported in the literature. All these schemes usually underestimate the band gap. A common correction of this underestimation is the scissors operator, which can be used even when some non-rigid shifts of bands occur in semiconductors, provided that the k dependence of the error in the excitation energies is negligible. In particular, this approximation is often used in the determination of the band gap offset in considering interfaces between different semiconductors, and also when optical transitions are studied. Even though there are formal theoretical improvements like the GW and BSE methods, these are still applied in solids using the k–independent scissors approximation due to its good performance and also to maintain the volume of calculations in a tractable way. Time–dependent DFT (TDDFT) (Ref. 56) is also making progress in non–linear optical properties such as those shown under the incidence of “strong” laser beams or ultrafast phenomena in semiconductors. We have not used the scissors operator since our electronic structure calculations reproduce quite well the experimental band structure and our band gap is comparable to those reported in the literature.

B. Self-consistent electronic structure

The calculation of optical properties begins with a self-consistent electronic structure. This is a key requisite since the calculation of optical properties involves matrix elements of the momentum operator, and the eigenfunctions used in their calculation must be precise. We used 120 irreducible k points in the BZ for the self-consistent calculation, with a product of 7. Here Rₖ is the smallest of all atomic sphere radii and Kmax is the plane-wave cutoff. The system has 28 (i.e., 7 times 4) atoms in the unit cell with seven crystallographically inequivalent sites (four indium and three selenium), each of which occupying four equivalent positions.

C. Optical properties

From our electronic structure calculations, we determined the imaginary part, ε₂(ω), of the complex dielectric function, ε(ω), integrating in k space (by the standard tetrahedron method). The general expression for the complex dielectric tensor is

\[ \varepsilon_2(\omega)_{\alpha\beta} = \frac{4\pi^2\varepsilon_0^2}{m^2} \sum_{i,j} \langle f_j | p_{\alpha} i | f_i \rangle \langle f_i | p_{\beta} i | f_j \rangle \]

\[ \times W_n (1 - W_f) \delta(E_f - E_i - \hbar \omega) d^3 k, \]

where \( \langle f_j | p_{\alpha} i | f_i \rangle \) and \( \langle f_i | p_{\beta} i | f_j \rangle \) are the dipole matrix elements corresponding to the α and β directions of the crystal (x, y, or z), and f and i are the final and initial states, respectively, \( W_n \) and \( E_n \) are the Fermi distribution function and

![Image](88x485 to 256x741)

**FIG. 1.** (Color online) Two unit cells of In₄Se₃. The axes x, y, and z correspond to the lattice parameters (Ref. 12) \( a = 15.297 \) Å, \( b = 12.308 \) Å, and \( c = 4.081 \) Å, respectively. The cleavage is perpendicular to axis x. The seven inequivalent atoms are labeled in the lower unit cell.
electron energy for the nth state, respectively. The real part of
the diagonal dielectric functions is computed from \(\varepsilon_2(\omega)\)
using the Kramers-Kronig relations in the form
\[
\varepsilon_1(\omega)_{aa} = 1 + \frac{2}{\pi} \int_{0}^{\infty} \frac{\omega \varepsilon_2(\omega')_{aa}}{\omega^2 - \omega'^2} d\omega',
\]
where \(P\) means the principal value of the integral. In order to
give a complete discussion of the optical spectra of In\(_2\)Se\(_3\),
we also studied the real and imaginary parts of the complex
refraction index \(n(\omega)_{aa} = \varepsilon_1(\omega)_{aa} + ik(\omega)_{aa}\), where \(n(\omega)\) is the
ordinary refraction index and \(n(\omega)\) is the coefficient of ex-
tinction, obtained from \(\varepsilon(\omega)_{aa} = \varepsilon_1(\omega)_{aa} + i\varepsilon_2(\omega)_{aa}\). Finally,
we also computed the reflectivity according to
\[
R(\omega) = \left|\frac{1 - n(\omega)_{aa}}{1 + n(\omega)_{aa}}\right|^2
\]
where * means conjugate complex, the coefficient of absorption,
\[
\alpha(\omega)_{aa} = \frac{2\omega}{c} \left( -\varepsilon_1(\omega)_{aa} + \frac{\varepsilon(\omega)_{aa}}{2} \right)^{1/2} = \frac{2\omega}{c} k(\omega)_{aa}
\]
and the conductivity using
\[
\sigma = \frac{\omega}{4\pi} \text{Im}[\varepsilon(\omega)_{aa}] = \frac{\omega}{4\pi} \varepsilon_2(\omega)_{aa}.
\]
Our calculations, based on a ground state scheme as DFT,
could imply inaccuracies due to the omission of the local-
field correction and the electron-hole interaction. In order to
improve these inaccuracies, new techniques capable of incor-
porating them to the absorption spectra description (still built
upon the DFT framework) are being developed.49,52,61–63

In spite of those inaccuracies we found good agreement be-
tween theory and experiment, which suggests that those non-
linear effects are not of importance of In\(_2\)Se\(_3\). This requires
further confirmation by further theoretical and experimental
studies. More sophisticated approaches to the theory than
DFT and extending the energy range of the experimental
measurements are likely to prove valuable.

III. EXPERIMENTAL DESCRIPTION AND ANALYSIS

Generalized ellipsometry allows for determination of the
optical constants or alternatively dielectric constants for bi-
axial (dielectrically anisotropic) materials. For arbitrarily an-
isotropic materials, the complex ratio \(\rho\) of the s- and
p-polarized reflectivities depends on the polarization state of the
incident light. Measurement of \(\rho\) can be addressed within
different presentations of the electromagnetic plane-wave re-
sponse. Here we made use of the Stokes descriptive system.
Real-valued Mueller matrix elements M\(_{ij}\) connect the Stokes
parameters before and after sample interaction.54,65 The
Stokes vector elements for the traditional p-s polarization
system are: \(S_0 = I_p + I_s\), \(S_1 = I_p - I_s\), \(S_2 = I_{45} - I_{-45}\), \(S_3 = I_{0} - I_{90}\),
where \(I_p\), \(I_s\), \(I_{45}\), \(I_{-45}\), \(I_{0}\), \(I_{90}\) denote the intensities for the p,
s, +45°, −45°, right-, and left-handed circularly polarized
light components, respectively. Data analysis requires non-
linear regression methods, where measured and calculated
generalized ellipsometry data were matched as closely as possible by varying appropriate physical model parameters.
The data analysis require the setup of models for geometry
(sample crystal axes orientation), and polarizability proper-
ties of the materials involved in the sample of interest.

For orthorhombic materials the major dielectric function
values \(\varepsilon_{xx}, \varepsilon_{yy}, \text{ and } \varepsilon_{zz}\) for polarization along crystal axes a,
b, and c, respectively, and Euler angle coordinates \(\varphi, \psi, \text{ and } \theta\)
provide a complete description of the dielectric function
tensor \(\varepsilon\) within the laboratory frame of reference, as defined
in Eq. (12) of Ref. 66 and in Fig. 1 of Ref. 67. In this work,
the so-called wavelength-by-wavelength data inversion
method was employed, i.e., in addition to model parameters
\(\varphi, \psi, \text{ and } \theta\) constant for all wavelengths, function values \(\varepsilon_{xx}, \varepsilon_{yy}, \text{ and } \varepsilon_{zz}\) were obtained at each wavelength independent
from spectrally adjacent wavelengths. Thereby, no a priori
physical line shape model was implemented into our model
analysis schemes, allowing for model unbiased extraction of
intrinsic dielectric function values \(\varepsilon_{xx}, \varepsilon_{yy}, \text{ and } \varepsilon_{zz}\).

The angle-resolved (angle of incidence \(\Phi_\delta\) and in-plane
rotation angle \(\varphi\)) spectroscopic Mueller matrix ellipsometry
measurements were performed using a commercial instru-
ment (M2000, J. A. Woollam Co., Inc.) within a spectral
range from 0.76 to 3.1 eV. A near (100) surface of single
crystal In\(_2\)Se\(_3\) with a-axis orientation approximately 7° off
away from the surface normal was measured. The ellipso-
meter was mounted on an automatic variable \(\Phi_\delta\) and sample
rotator \(\varphi\) stage. \(\Phi_\delta\) was varied from 45° to 75° in steps of
10°, while \(\varphi\) was varied from 0° to 360° in steps of 5°. The
polarizer-compensator-sample-analyzer ellipsometer is capa-
bale of measuring 11 out of 16 Mueller matrix elements
normalized to \(M_{11}\) (except for elements in fourth row).54

Figures 2 and 3 depict experimental and best-match model
calculated data at selected wavelengths, angles of incidence
and sample azimuth rotations. Given the fact that In\(_2\)Se\(_3\) is a
corrugated but otherwise layered compound with an easy
cleavage parallel to (100) planes (i.e., perpendicular to the
a-axis), there are difficulties in preparing a high quality sur-
face parallel to the a-axis, i.e., \(\langle 100 \rangle\) direction or cleavage
plane normal. These, in turn, result in off axis ellipsometry
data insufficiently reliable for comparison of the experiment
with theory for \(\varepsilon_{xx}\). We have therefore restricted our com-
parison between experiment and theory for the real and
imaginary parts of the dielectric functions extracted along the
\(\langle 010 \rangle\) and \(\langle 001 \rangle\) directions (i.e., \(\varepsilon_{yy}\) and \(\varepsilon_{zz}\)) which we
discuss in Sec. IV B.

IV. DISCUSSION OF RESULTS

A. Band structure and DOS

Figure 4 shows the energy band structure with spin-orbit
(SO) interaction—which is almost identical to those without,
while Figs. 5–7 show several DOS.

The electronic structure of In\(_2\)Se\(_3\) without spin–orbit can
be understood as four main bundles of valence bands and
two of conduction bands (see Fig. 4 and specially bottom
floor of Fig. 5). There is a flat isotropic band between −15.30
and −14.25 eV, mostly due to the In d orbitals and a much
lesser contribution of Se s states. This band is followed by a
less populated band resulting from Se $s$ but also from the In $d$ orbitals (though with a less significant contribution) between $-13.50$ and $-11.90$ eV binding energies. Then there is an empty (unoccupied) region between $-11.90$ and $-6.83$ eV below the Fermi level, where the bottom of the valence band begins. At the bottom of the valence, we find four noticeable peaks (A–D in Fig. 5) due to similar contributions from In $s$ and Se $p$ electrons. The bands between $-5.26$ and $-4.04$ eV have equal contributions from In $s$ and Se $p$ orbitals. The bands above $-4.04$ eV are mostly formed by Se $p$ electrons. As general observation, we note that crystallographically each of the nonequivalent Se atoms contribute an almost identical density of state (see second panel of Fig. 5). In contrast, In atoms show some differences, especially In4 atoms (see peaks C$_{In}$ and F$_{In}$ in the top panel of Fig. 5) with respect to the other In atoms.

![FIG. 2.](image1) (Color online) Selected experimental (symbols) and best-match model calculated GE data $M_{13}$, $M_{14}$, $M_{23}$, $M_{24}$ versus sample azimuth rotation, at photon energy 2.1 eV, and various angle of incidence ($\Psi_{a}=45^\circ$,$55^\circ$, $65^\circ$,$75^\circ$).

![FIG. 3.](image2) (Color online) Selected experimental (symbols) and best-match model calculated GE data $M_{13}$, $M_{14}$, $M_{23}$, $M_{24}$ versus photon energy at sample azimuth rotation $240^\circ$, and angle of incidence ($\Psi_{a}=45^\circ$, $55^\circ$, $65^\circ$, $75^\circ$).

![FIG. 4.](image3) Band structure with the inclusion of the spin orbit interaction, calculated with the Engel–Vosko GGA.

![FIG. 5.](image4) (Color online) Total DOS of the seven inequivalent atoms of the unit cell and (see bottom floor) comparison of the total DOS for the whole system with against without the inclusion of spin-orbit. wso: with SO, woso: without SO.
The conduction band is mainly $p$-like (with both In and Se contributions). However, there is a sharp peak in the DOS due to the In $s$ orbitals near the bottom of the conduction band. A similar situation holds for the narrow In4 $4s$ states near the valence band maximum. These $s$-cation states, hybridized with their $p$-cation and -anion counterparts, are very important for the physical properties of this material, since they define the band gap. The band gaps with and without spin–orbit are almost the same: 0.57 and 0.58 eV, respectively, both of them direct at $\Gamma$. Further, the morphology of the bands with and without spin–orbit is almost identical (see also comparison of the DOS in the bottom panel of Fig. 5), except for minor splittings of the band all over the BZ. There is however a strong and noticeable splitting of the bands around $\pm 15$ eV into four sharply defined bands. Table II shows the approximate location of the main structures of the total DOS, as well as the most important contributions to them, atomic and orbital resolved.

### Table II. Approximated location (E) of main total DOS structures (labeled A to H, bottom floor of Fig. 5) and its component, atom- and orbital-resolved.

<table>
<thead>
<tr>
<th>E (eV)</th>
<th>Main atomic contributions</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$-6.35$ A$_{4s}$ (In2 $s$, In1 $s$, In3 $s$)</td>
</tr>
<tr>
<td>B</td>
<td>$-5.69$ B$_{4s}$ (In5 $s$, In1 $s$, In4 $s$)</td>
</tr>
<tr>
<td>C</td>
<td>$-5.07$ C$_{4s}$ (In1 $s$)</td>
</tr>
<tr>
<td>D</td>
<td>$-4.44$ D$_{4s}$ (In4 $s$, In2 $s$, In3 $s$, In1 $s$)</td>
</tr>
<tr>
<td>E</td>
<td>$-2.49$ E$_{4s}$ (Se $p$)</td>
</tr>
<tr>
<td>F</td>
<td>$-1.31$ F$_{4s}$ (Se $p$)</td>
</tr>
<tr>
<td>G</td>
<td>$-0.67$ G$_{4s}$ (In4 $s$, In2 $s$, In1 $s$, In3 $s$)</td>
</tr>
<tr>
<td>H</td>
<td>$1.64$ H$_{4s}$ (In1 $s$, In2 $s$, In3 $s$, In4 $s$)</td>
</tr>
</tbody>
</table>

B. Optical properties

In order to assess convergence of the optical properties, we calculated $\varepsilon_2$ with increasingly finer meshes for the discretization of the BZ: 144, 192, and 300 $k$ points without the spin-orbit (SO) interaction, and 144 $k$ points with it. All of them were coincident, therefore, we assume convergence at 144 $k$ points and report all optical spectra with spin orbit. All the calculated optical spectra are presented in Fig. 8 ($\varepsilon_1$, $\varepsilon_2$, $n$, and $k$) and Fig. 9 ($R$, $\alpha$, and $\sigma$), and the location (in energy) of their main structures is tabulated in Tables III and IV. It can be seen in all spectra that the material has a triaxial anisotropy for energies up to $\sim 1.5$ eV, and a quasi-2D behavior for higher energies, since curves for the $x$ and $y$ axes are very similar (though not identical), while those for the $z$ axis are clearly different (in amplitude and/or shape). Finally, we note that isotropy only appears in small isolated energy ranges above 15 eV.

Dispersion. The real part of the dielectric function ($\varepsilon_1$) is closely related to the dispersion of incident photons by the material, so is the refractive index ($n$), and in a less obvious way is the reflectivity (that actually results from the combination of absorption and dispersion), which are presented in Fig. 8 ($\varepsilon_1$ and $n$) and 9 ($R$). It can be seen that $n_{xx}$ has its main peak (1) coinciding with that of $n_{yy}$ (0) at 1.84 eV. In
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FIG. 8. (Color online) Top to bottom floors: real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) part of the dielectric function comparing with experiment for $yy$ and $zz$ axes, index of refraction ($n$) and coefficient of extinction ($k$). The position (in eV) of the labeled peaks is in Table III.

In fact both indexes coincide almost exactly in the range from $\sim 1.3$ to 3.0 eV and also for $E \sim 3.5$ eV, i.e., the material is birefringent in the near IR-visible and also from the UVB up in the spectrum. It shows, however, a triaxial behavior in the range 3.0–3.5 eV (blue to UVA range). As to $n_{zz}$, it shows a blunt plateau rather than a pronounced peak. The real part of $\varepsilon$ follows closely the morphology of $n$, and for $yy$ and $zz$ axes we also present ellipsometry measurements showing excellent agreement with theory for energies up to $\sim 2.0$ eV. While there is a faster drop of the remaining experimental points for higher energies, up to $3.1$. With respect to the reflectivity, it oscillates between $\sim 30\%$ and $\sim 50\%$ for incident energies up to $\sim 12.5$ eV, where it drops sensibly. In addition, it is to be noticed that reflectivity shows strong anisotropy for energies above $7.5$ eV while in the other properties, anisotropy apparently decreases: reflectivity just enhances the effect of what in other properties appears as small differences among the different axes.

Energy intake. The imaginary part of the dielectric function ($\varepsilon_2$) is closely related to the intake of energy by the material, and so are the coefficients of extinction ($k$) and absorption ($\alpha$), and the optical conductivity ($\sigma$), which are

<table>
<thead>
<tr>
<th>Peaks</th>
<th>$e_1$</th>
<th>$e_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E=0$</td>
<td>13.04</td>
<td>11.61</td>
</tr>
<tr>
<td>0</td>
<td>0.70</td>
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<td>1</td>
<td>1.75</td>
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<tr>
<td>2</td>
<td>3.50</td>
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<tr>
<td>3</td>
<td>4.85</td>
<td>5.19</td>
</tr>
<tr>
<td>4</td>
<td>9.13</td>
<td>6.71</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Peaks</th>
<th>$n$</th>
<th>$k$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E=0$</td>
<td>3.62</td>
<td>3.41</td>
</tr>
<tr>
<td>0</td>
<td>0.69</td>
<td>1.91</td>
</tr>
<tr>
<td>1</td>
<td>1.84</td>
<td>3.06</td>
</tr>
<tr>
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<td>3.61</td>
<td>5.76</td>
</tr>
<tr>
<td>3</td>
<td>4.94</td>
<td>6.69</td>
</tr>
<tr>
<td>4</td>
<td>9.02</td>
<td>9.41</td>
</tr>
</tbody>
</table>
TABLE IV. Values at E=0 and approximate positions (in eV) of main peaks of the optical properties of In$_4$Se$_3$.

<table>
<thead>
<tr>
<th>Peaks</th>
<th>$\alpha$</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th>Re[\sigma]</th>
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<tbody>
<tr>
<td></td>
<td>$x$</td>
<td>$y$</td>
<td>$z$</td>
<td>$x$</td>
<td>$y$</td>
<td>$z$</td>
<td>$x$</td>
<td>$y$</td>
<td>$z$</td>
<td></td>
</tr>
<tr>
<td>$R_{G=0}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>1</td>
<td>0.72</td>
<td>1.10</td>
<td>1.30</td>
<td>0.32</td>
<td>0.30</td>
<td>0.27</td>
<td>0.64</td>
<td>1.16</td>
<td>1.51</td>
<td></td>
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<tr>
<td>2</td>
<td>2.98</td>
<td>3.29</td>
<td>5.15</td>
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<td>4.10</td>
<td>3.85</td>
<td>3.27</td>
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</tr>
<tr>
<td>3</td>
<td>3.90</td>
<td>4.03</td>
<td>6.21</td>
<td>4.49</td>
<td>5.11</td>
<td>5.11</td>
<td>4.25</td>
<td>4.00</td>
<td>5.84</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>5.60</td>
<td>7.34</td>
<td></td>
<td>5.47</td>
<td>7.18</td>
<td>6.16</td>
<td>5.24</td>
<td>4.44</td>
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<td></td>
</tr>
<tr>
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<td>6.53</td>
<td>8.61</td>
<td>7.69</td>
<td>6.33</td>
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<td>8.30</td>
<td>7.80</td>
<td>5.44</td>
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<td>6</td>
<td>7.85</td>
<td>9.84</td>
<td>9.36</td>
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<td></td>
<td>9.07</td>
<td>7.00</td>
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</tr>
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</tr>
<tr>
<td>8</td>
<td>11.35</td>
<td>12.40</td>
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V. CONCLUSIONS

We have performed what to the best of our knowledge is the first computational analysis of the optical properties of the binary compound In$_4$Se$_3$. We also performed ellipsometry measurements and determined the real and imaginary parts of the dielectric function, finding good agreement with the theoretical results. Given the scarce availability of experimental measurements on the system, our spectra result rather predictive, showing bi- and triaxial anisotropy for different ranges of incident photon energy. We highlight the importance of our calculations for the $xx$ axis, since measurements in that direction face serious experimental difficulties.

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