Optical properties and electronic structures of the intermetallic compounds AuGa$_2$ and PtGa$_2$

Kwang-Joo Kim  
*Iowa State University*

Bruce N. Harmon  
*Iowa State University*, harmon@ameslab.gov

Liang-Yao Chen  
*Iowa State University*

David W. Lynch  
*Iowa State University*, dlynch@iastate.edu

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Abstract
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Keywords
Ames Laboratory, spectroscopic ellipsometry, electric dipole matrix elements

Disciplines
Atomic, Molecular and Optical Physics | Condensed Matter Physics | Physics

Comments
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Optical properties and electronic structures of the intermetallic compounds AuGa$_2$ and PtGa$_2$

Kwang Joo Kim, B. N. Harmon, Liang-Yao Chen,* and David W. Lynch

Department of Physics and Ames Laboratory, U.S. Department of Energy, Iowa State University, Ames, Iowa 50011

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The electronic structures of AuGa$_2$ and PtGa$_2$ have been studied with use of spectroscopic ellipsometry and the dielectric functions have been determined in the 1.2–5.5-eV region. Both compounds show interband absorption at low photon energies (< 1.3 eV). The interband absorption for AuGa$_2$ is strong at about 2 eV while that for PtGa$_2$ shows a broad structure in the range 2.5–4.5 eV, with a shoulder at 3.3 eV. The observed interband features in $\epsilon_2$ can be interpreted in terms of self-consistent relativistic band-structure calculations using the linear augmented-plane-wave method. The interband contribution to the imaginary part of the dielectric function $\epsilon_2^I$ has been calculated including the effect of the electric dipole matrix elements. The overall agreement is good between the band calculations and the ellipsometry results in the 1.2–5.5-eV region for both the magnitude and the positions of the structures. Below 1.2 eV the calculational results for both compounds show interband absorption, which is also qualitatively suggested by the ellipsometry results.

I. INTRODUCTION

The binary intermetallic compounds AuGa$_2$ and PtGa$_2$ have the cubic fluorite (CaF$_2$) structure in which Au or Pt atoms form a fcc sublattice and the Ga atoms occupy the tetrahedral sites located one-quarter of the way up the body diagonals. Earlier studies of the electrical properties of AuGa$_2$, AuAl$_2$, and AuIn$_2$ (Ref. 1) showed that they are good metallic conductors with room-temperature conductivities about one-fifth that of Cu. Also, Fermi-surface studies on these compounds$^{2,3}$ indicated that they behave nearly-free-electron-like to some extent. Optical reflectivities were measured on these compounds$^{4,5}$ and dielectric functions were calculated by using Kramers-Kronig (KK) analyses. Chen and Lynch$^6$ measured the complex dielectric functions of AuAl$_2$ and PtAl$_2$ by spectroscopic ellipsometry. Using published band structures of the compounds$^7$ they were able to assign some interband transitions to the structures observed in the dielectric functions.

As is well known, the electronic band structure of Au (Refs. 8–10) is characterized by narrow 5$d$ bands which hybridize significantly with the $s$-$p$ bands around the Brillouin-zone boundary. Also, the spin-orbit interaction splits the 5$d$ states into states with $j = \frac{3}{2}$ and $j = \frac{1}{2}$. This ordering combines with crystal-field effects so that at the $\Gamma$ point, the spin-orbit and crystal-field interactions cause the fourfold $j = \frac{3}{2}$ states (upper $\Gamma_8^+$) to shift to higher energy and the $j = \frac{1}{2}$ states to split into fourfold (lower $\Gamma_8^+$) and twofold ($\Gamma_7^+$) levels. The spin-orbit interaction enhances the $d$-band width, and the energy gap between the $j = \frac{3}{2}$ levels at the $\Gamma$ point is about 1 eV for Au, a gap large enough to be detected in photoemission measurements.$^{11,12}$

In AuGa$_2$, according to the earlier band calculations on this compound,$^{11,13}$ the width of the Au-derived 5$d$ bands is narrower than that of elemental Au because by forming a compound with Ga the distances between Au atoms become approximately 1.5 times larger than in elemental Au which results in weaker $d$-$d$ overlap interactions between Au atoms. Also, the lowest-lying $s$ band reaches the zone boundary (e.g., $X_1$) without crossing the $d$ bands. This results in less $s$-$d$ hybridization and the $d$ bands reside within a band gap between $s$ and $p$ bands. Therefore, in AuGa$_2$, the electronic states near the Fermi level $E_F$ are expected to have mostly $s$-$p$ character of both constituents and the interband transitions between them are expected to contribute to the low-energy (< 5 eV) optical properties of the compound. This is in contrast to the case for pure Au which has interband contributions below 5 eV involving the $d$ bands$^{14,15}$ which extend higher in energy compared to those of AuGa$_2$. The electronic properties of PtGa$_2$,$^{16}$ on the other hand, are not so well studied as those of AuGa$_2$. However, we can at least estimate the qualitative differences in the electronic structure between PtGa$_2$ and Pt based on those between AuGa$_2$ and Au.

The electronic structure information gained from optical measurements comes from the spectral dependence of the optical response described by the complex dielectric
function which involves the characteristic energies of prominent excitations, the joint density of states, and the relative oscillator strengths of the excitations. The complex dielectric function of a metal can be described as the sum of an intraband term and an interband term,
\[ \varepsilon = \varepsilon^i + \varepsilon^b. \]

The intraband term of the optical spectra can be treated quantitatively within the framework of the Drude model as
\[ \varepsilon_i = 1 - \frac{\omega_p^2 \tau^2}{1 + \omega^2 \tau^2}, \]
\[ \varepsilon_i^b = \frac{\omega_p^2 \tau}{\omega(1 + \omega^2 \tau^2)}. \]

The two parameters of the model are \( \omega_p^2 \) (= \( 4\pi Ne^2/m^* \)), the bulk plasma frequency squared, and \( \tau^{-1} \), the electron scattering rate. The interband term depends upon the details of the electronic band structure. It is related to momentum-conserving electric dipole transitions between states separated by the photon energy.

In this work, we report ellipsometric measurements of the dielectric functions of AuGa\(_2\) and PtGa\(_2\) in the 1.2–5.5-eV region which we interpret in terms of intraband and interband transitions. We also performed self-consistent band calculations for both compounds using the linearized augmented-plane-wave (LAPW) method. The calculations were done semirelativistically including the spin-orbit interaction as a perturbation. Using the calculated one-electron eigenfunctions, we calculated the electric dipole matrix elements for the direct interband transitions between the occupied \( |i\rangle \) and the unoccupied \( |f\rangle \) states \( P_{fi} \) given by
\[ P_{fi} = \frac{\hbar}{\Omega} \langle f | \nabla | i \rangle \]
inTEGRATED over the unit cell of volume \( \Omega \). These are related to the interband contribution to the imaginary part of the complex dielectric function \( \varepsilon_b^b \) by
\[ \varepsilon_b^b = \frac{4\pi^2 e^2}{3m^2 \omega^2} \sum_{f,i} \int_{BZ} d^3 k \left| \frac{2}{(2\pi)^3} |P_{fi}|^2 \delta(E_f - E_i - \hbar \omega) \right| , \]
which is to be compared with the experimental results. We did not include the spin-flip terms of the relativistic matrix elements as they are expected to be small.

II. EXPERIMENTAL

The raw materials used in this work were bulk Au, Pt, and Ga, all 99.99% purity. The AuGa\(_2\) sample is a polycrystalline made by arc melting in an argon atmosphere. The weight loss during melting did not exceed 0.1%. The PtGa\(_2\) sample is a single crystal which is stable at room temperature. It was grown by the Bridgman method which is described in detail elsewhere, oriented using Laue x-ray diffractometry, then cut to reveal a (111) surface. The AuGa\(_2\) sample has a slight bluish hue and the PtGa\(_2\) sample is a yellow color like that of gold.

The lattice parameters obtained by x-ray-diffraction measurements are 6.036 Å for AuGa\(_2\) and 5.911 Å for PtGa\(_2\), which are in good agreement with the values in the literature. Before the measurement, the samples were mechanically polished with abrasives, the final grade being a paste of 0.05-μm-diam alumina, and afterwards cleaned by acetone and methanol.

The scanning photometric ellipsometer with rotating polarizer and analyzer, which has been discussed in detail previously, was used to determine the dielectric functions of the samples at room temperature. Measurements were made in the 1.2–5.5-eV range at energy intervals of 0.01 eV in the 1.2–2.5-eV range and 0.02 eV in the 2.5–5.5-eV range. The spectral bandpass was 3.3 nm and the measurement errors did not exceed 1% in \( \varepsilon_3 \). Systematic errors caused by oxide overlayer formation appear to be small. Such errors can cause a small shift in the absolute values of the spectra but do not alter the positions of the spectral features.

III. BAND-CALCULATIONAL DETAILS

The crystal structure of AuGa\(_2\) and PtGa\(_2\) is cubic fluoride (CaF\(_2\)). The Bravais lattice is fcc, the space group is \( O_h^2 (Fm\overline{3}m) \), and this structure has three atoms per unit cell. The lattice constants are 6.06 Å for AuGa\(_2\) and 5.91 Å for PtGa\(_2\).

Electronic band-structure calculations for both compounds were performed using the LAPW method, employing the standard muffin-tin approximation for the crystal potential, which provides a good approximation for a cubic close-packed structure. The calculations were scalar relativistic, in which the Dirac equation is reduced to omit initially the spin-orbit interaction (thus keeping spin as a good quantum number but retaining all other relativistic kinematic effects). The spin-orbit interaction is added perturbatively after the semirelativistic bands and wave functions have been obtained.

The starting muffin-tin crystal potentials were constructed from the superposition of neutral atomic charge densities obtained from self-consistent atomic calculations by the Dirac-Slater method using a variation of the Liberman-Waber-Cromer program, which includes relativistic effects. The atomic configurations of Au and Ga were \( 5d^{10}6s^1 \) and \( 4s^24p^1 \), respectively, in AuGa\(_2\) and those of Pt and Ga were \( 5d^96s^1 \) and \( 4s^24p^1 \), respectively, in PtGa\(_2\). The exchange-correlation contribution to the potential was calculated by using the local-density approximation of Hedin and Lundqvist to the density-functional formalism. This local-density potential has the advantage of being self-consistent and containing no adjustable parameters and has proved successful in accounting for the ground-state properties of a variety of metals. The crystal charge density was calculated from the wave functions of the filled states obtained by solving the Hamiltonian containing the effective crystal potential. By mixing the new charge density with the old charge density we can construct the new potential from this mixture and the self-consistency procedure was continued until the change in the charge in each muffin-tin sphere converged to within \( 10^{-4} \) electron and the energy eigenvalues to within \( 10^{-4} \) Ry in successive iterations.
The muffin-tin sphere radii were chosen so that the spheres surrounding the Au and Pt sites extended 57% and 56%, respectively, of the way up to the Ga site. With this arrangement only 47% of the unit-cell volume is occupied by the three muffin-tin spheres for both compounds. In other words, the crystal potential is taken as constant over 53% of the unit-cell volume. This is a questionable approximation to the real crystal potential. This problem of approximating the potential as a constant in a large region can be alleviated by including more of the unit-cell volume inside an additional sphere. This is accomplished by considering one additional sphere at the unoccupied octahedral site (one-half of the way up the body diagonal and between the Ga atoms). The size of this sphere is taken to be as large as possible without changing the sizes of the other spheres. The radii of the spheres are listed in Table I. By including this sphere the amount of the unit-cell volume enclosed within the muffin-tin sphere becomes 70% for AuGa$_2$ and 71% for PtGa$_2$. Although no nucleus is at the center of the added sphere, upon self-consistency it contains 0.70 electron for AuGa$_2$ and 0.66 electron for PtGa$_2$.

The size of the LAPW basis-function set for each calculation was set to satisfy $R_{\text{MT}} = 7.5$ for the smallest muffin-tin radius $R_{\text{MT}}$. This yielded about 170–200 LAPW’s for both compounds. Inside $R_{\text{MT}}$, the wave functions were expanded in terms of spherical harmonics of angular momentum up to $l = 12$. There were 46 $k$ points in the 24th of the Brillouin zone included for the self-consistent iterations.

We also calculated the density of states (DOS) for both compounds. We used the energy eigenvalues for the first 13 bands at 152 $k$ points. They were least-squares-fitted with 81 symmetrized plane waves (the typical rms error of the fits was less than 1 mRy). These fits were used to generate the band energies at the corners of 2048 small tetrahedra which filled the irreducible wedge (1/24) of the Brillouin zone. The DOS was then obtained using the linear-energy-tetrahedron method$^{27}$ which was also used to calculate the interband contribution to the imaginary part of the dielectric function $\varepsilon_2$. In calculating $\varepsilon_2$ as in Eq. (5), we needed to compute the electric dipole matrix elements as in Eq. (4). The energy eigenvalues were evaluated at the four corners of 110 elementary tetrahedra in the irreducible 24th of the Brillouin zone and the electric dipole matrix elements were calculated using the wave functions at the centers of the tetrahedra. We assumed the electric dipole matrix elements to be constant within any one tetrahedron, and equal to the matrix element calculated at the center of the tetrahedron.

### Table I. Parameters used in the band calculations.

<table>
<thead>
<tr>
<th></th>
<th>AuGa$_2$</th>
<th>PtGa$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lattice constant (Å)</td>
<td>6.06</td>
<td>5.91</td>
</tr>
<tr>
<td>Muffin-tin radius (Å)</td>
<td>Au 1.50</td>
<td>Pt 1.43</td>
</tr>
<tr>
<td></td>
<td>Ga 1.13</td>
<td>Ga 1.13</td>
</tr>
</tbody>
</table>

**FIG. 1.** Real and imaginary parts of the complex dielectric functions of AuGa$_2$ and PtGa$_2$.
energies for both compounds. This assumption has been a good approximation for noble metals. Then we can obtain the free-electron (Drude) parameters for both compounds which can be used to estimate the intraband contributions at higher energies.

We obtained the dielectric functions at low energies (<1 eV) by KK analyses of the reflectivity data in the 0.02–6.2-eV range measured by Wieliczka et al. at room temperature for both compounds. For energies larger than 6.2 eV and up to 100 eV reflectivities were assumed to drop off as \( \omega^{-4} \), and for still higher energies as \( \omega^{-4} \), where \( p \approx 4 \) is an adjustable parameter to make the calculated dielectric functions agree in magnitude with the ellipsometry data in the 1.2–5.5-eV region. The structure of the spectra is not sensitive to values of \( p \).

In estimating the Drude parameters we assume that \( \varepsilon \approx \varepsilon' \) for the dielectric functions at low energies (<0.5 eV) obtained from KK analyses of the reflectivity data for both compounds, which means we neglect the contributions from interband absorption at low energies. We also assume that \( \omega \tau \gg 1 \) at the same energy range which has been a good approximation for noble metals. For AuGa\(_2\), the dc relaxation time \( \tau_{dc} \) was estimated to be \( 8.0 \times 10^{-14} \) sec from the dc resistivity, so that the above condition is satisfied at 0.5 eV. Then Eqs. (2) and (3) reduce, respectively, to

\[
\varepsilon_1 = \varepsilon_\infty - \frac{\omega_\text{p}^2}{\omega^2},
\]

\[
\varepsilon_2 = \frac{\omega_\text{p}^2}{\omega^2 \tau},
\]

where \( \varepsilon_\infty = 1 + \varepsilon_\text{b} \) with \( \varepsilon_\text{b} \) the contribution to \( \varepsilon_1 \) from interband transitions at higher energies.

By linear-regression fitting of the real part of the dielectric functions below 0.5 eV we can obtain \( \varepsilon_\infty \) and \( \omega_\text{p} \). Also, it is known that in order to fit the low-energy optical spectra for a variety of metals \( \tau \) needs to be frequency dependent.\(^{30,31}\) For the noble metals, it is empirically given by\(^{32,33}\)

\[
\tau^{-1} = \tau_0^{-1} + \beta \omega^2,
\]

and we used this relation to fit the imaginary parts of the dielectric functions below 0.5 eV to get \( \tau_0 \) and \( \beta \). The parameters obtained from the above procedure are listed in Table II.

We used the above four-parameter sets for both compounds to estimate the intraband parts of the dielectric functions at higher energies (>1.2 eV). The intraband parts were subtracted from the ellipsometry data to get the interband parts. The \( \varepsilon_2^\ast \) for PtGa\(_2\) obtained from the above procedure shows the structure at about 1.3 eV more developed than in Fig. 1. However, the \( \varepsilon_2^\ast \)'s for both compounds still show increasing low-energy ends which might mean that there are strong interband transitions below 1.2 eV, the low-energy limit of our measurements. For this reason, the Drude parameters may not be meaningful for both compounds. They are used only to try to separate \( \varepsilon \) into two parts.

### B. Band structures and optical transitions

Relativistic band structures of AuGa\(_2\) and PtGa\(_2\), including the spin-orbit interaction are shown in Figs. 3 and 4 along certain high-symmetry directions. In both structures, the bands near \( E_F \) are of Ga \( s-p \) character mixed with Au or Pt \( s-p \) character and these \( s-p \) bands along the \( \Gamma-K \) and \( L-W \) directions have quite similar shapes in the two materials except for the difference in \( E_F \). Also, these bands do not shift much in energy by including the spin-orbit interaction as a perturbation while the lower-lying \( d \) bands shift and split significantly due to the combined spin-orbit and crystal-field effect.

The lowest band for both compounds consists of a mixture of Au 6s– or Pt 6s–Ga 4s bonding states. Its hybridization with 5d states of Au or Pt is much weaker than those in elemental Au or Pt. The narrow set of bands above this \( s \) band are 5d bands of Au or Pt. The positions of these 5d bands in AuGa\(_2\) agree within 0.5 eV with the result of the angle-resolved photoemission measurement on the (100) surface of AuGa\(_2\).\(^{34}\) Also, the measured energy gap between \( \Gamma_7 \) and the lower \( \Gamma_7' \) from the above measurement is 1.6 eV while our calculation gives 1.1 eV. In PtGa\(_2\), these 5d bands are located at higher energies than in AuGa\(_2\); the upper \( \Gamma_7' \) is located 2.3 eV higher in PtGa\(_2\) than in AuGa\(_2\) relative to \( E_F \). In Table III a comparison is made between the 5d states at \( \Gamma \) with and without the spin-orbit interaction for both compounds. It is seen that the widths of the 5d bands are increased due to the rise of the doubly degenerate \( \Gamma_7(\varepsilon_8) \) level to \( \Gamma_7^* \) and also due to the splitting of the triply degenerate \( \Gamma_{25}(t_{2g}) \) level into the twofold \( \Gamma_7' \) and fourfold \( \Gamma_8' \). The width of the 5d bands estimated from the DOS

![Graph](image-url)

**FIG. 2.** Normal-incidence reflectivity spectra of AuGa\(_2\) and PtGa\(_2\). Note that the zero of the reflectivity is suppressed.
of AuGa$_2$ in Fig. 3 agrees well with the result of the valence-band x-ray photoemission spectroscopy (XPS) measurement.\cite{15,16}

The main difference between the band structures of AuGa$_2$ and PtGa$_2$ near $E_F$ is that for AuGa$_2$ the band originating from the $\Gamma$-point is located below $E_F$ along the $\Gamma-X$, $\Gamma-K$, and $\Gamma-L$ directions while for PtGa$_2$ it is above $E_F$ along the $\Gamma-X$ direction and partially unfilled along the $\Gamma-K$ and $\Gamma-L$ directions. This band is derived mostly from Ga 4$s$ antibonding states among neighboring Ga atoms. In AuGa$_2$, this band is quite flat along the $\Gamma-X$ direction and located 0.26 eV below $E_F$ at the $\Gamma$ point. However, this band does not cause a high density of states at about 0.26 eV because it disperses significantly in the other directions of the Brillouin zone. In other calculations,\cite{7,11} this band lies at lower energies and at the $\Gamma$ point it is located about 1.4 eV below $E_F$, differing from our result by more than 1 eV.

We calculated the interband contribution to the imaginary part of the dielectric function $\varepsilon''_2$ for both AuGa$_2$ and PtGa$_2$ according to Eq. (5), including the effect of the electric dipole matrix elements. In Figs. 5 and 6 we compare our calculation results with the spectra obtained after subtracting the Drude-model calculations from the optical data. We can see that overall agreement is good between the two quantities for both the peak positions and the strengths of the structures for both compounds in our ellipsometry data range (1.2–5.5 eV). An interesting result is that both compounds show strong calculated interband absorptions below our ellipsometry data range: for AuGa$_2$ there is a peak at 0.65 eV, and for PtGa$_2$ there is a peak at 0.97 eV. At 1.2 eV, the strength of this interband absorption compared to the total absorption strength $\varepsilon''_2$ is 86% for AuGa$_2$ and 66% for PtGa$_2$. The reflectivity data of Wieliczka et al.\cite{29} for both compounds do not show any structure at energies below 1 eV so that the dielectric functions obtained from them do not show any structure related to the interband absorptions in this range. This is possible if the strengths of the

![Figure 3](image1.png)

FIG. 3. Relativistic energy-band structure and density of states of AuGa$_2$ including spin-orbit coupling.

![Figure 4](image2.png)

FIG. 4. Relativistic energy-band structure and density of states of PtGa$_2$ including spin-orbit coupling.

![Figure 5](image3.png)

FIG. 5. Comparison of the interband contribution to the imaginary part of the complex dielectric function for the band calculation (broadened by convolution with a Lorentzian of full width at half maximum of 0.05 eV) and the experiment for AuGa$_2$.
intraband absorptions at room temperature are much greater than those of the interband absorptions so that the reflectivity measurements could not resolve the weaker interband contributions from the stronger intraband contributions. Another possibility is that our calculations might overestimate the strengths of the low-energy interband absorptions, or place them slightly too high in energy, where they would appear to be more observable. By calculating the interband contribution to $\epsilon_1$ from our calculated interband $\epsilon_2$, adding these to the Drude $\epsilon$, and calculating the reflectivity, we find no detectable structure in reflectivity is expected in PtGa$_2$ due to the low strength of these interband transitions. Such a structure should be detectable in AuGa$_2$ if the calculations are accurate.

As a good example, the reflectivity measurement on Al (Ref. 37) at room temperature could not resolve a structure due to the interband absorption at low energies ($<1$ eV) from the background of intraband absorption. Also, KK analysis$^{39}$ using various room-temperature optical data, including the above, could not produce the low-energy structure. However, an absorptivity measurement on Al at 4.2 K (Ref. 39) showed an interband absorption peaked at about 0.5 eV. Therefore, for some metals, phonon-assisted intraband absorption might increase dramatically as the temperature rises so that at room temperatures we might not resolve the interband absorptions at low energies.

To find the regions in k space which contribute significantly to the structures in $\epsilon_1$ we calculated the integral part of Eq. (5) for each of the 110 tetrahedra for selected energy windows centered at about the peaks of each structure for both compounds. The intensities of all the interband transitions having energy differences between the initial and final states falling in the energy window were summed for each tetrahedron to represent the contribution from the tetrahedron to the structure. In Figs. 7 and 8 each tetrahedron is represented by a rectangle if it makes a contribution to the structure, with the size of the rectangle denoting the strength of the contribution. Figure 7 shows the distribution of the transitions for the 0.65-eV peak of AuGa$_2$ in the $\Delta$th of the Brillouin zone. The largest contribution comes from around the middle of the $\Gamma-X$ line. These transitions occur between bands having mostly s-p character. As shown in Fig. 3 there are three nearly-parallel bands along the $\Gamma-X$ direction. The lowest band of the three at the X point (band 8) has Au s-p character and the other two upper bands (bands 9 and 10, respectively) have mixtures of Au p and Ga p character at around the middle of the $\Gamma-X$ line. The 0.65-eV peak is caused primarily by transitions between bands 8 and 9. At about 1 eV the strength of the transitions between bands 8 and 10 is comparable to that
the energy increases to make a shoulder at about 1.3 eV in $e_2^b$ of PtGa$_2$. The contributions to the 2-eV structure of AuGa$_2$ are throughout nearly all of the irreducible wedge and the contribution from the high-symmetry regions is very small compared to the total strength, as can be seen in Fig. 7. These contributions come mainly from transitions between filled bands having mixtures of Au $p$ and Ga $p$ character (like bands 9 and 10 along the $\Gamma-X$ direction) and unfilled bands having Ga $s-p$ character. Figure 8 shows the contributions to the 3-eV structure of PtGa$_2$. The contributions occur throughout all of the irreducible wedge. These transitions are mostly between the filled bands with Pt $s-p$ character and the unfilled bands with Pt $p-d$ character mixed with Ga $p$ character.

V. CONCLUSIONS

The optical properties of intermetallic compounds AuGa$_2$ and PtGa$_2$ in the 1.2–5.5-eV region agree well with the results from relativistic band-structure calculations including the spin-orbit interaction in the absolute magnitudes as well as the positions of the structures. Below 1.2 eV the calculational results for both compounds exhibit peaks which are due to transitions involving unfilled bands having Ga $p$ character. The interband terms $e_2^b$ for both compounds also show rising trends at low energies which may be evidence for the existence of the above-mentioned transitions below 1.2 eV as in the case of Al.

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*Present address: Department of Electrical Engineering, University of Nebraska–Lincoln, Lincoln, NE 68588-0511.