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# Ellipsometric study of optical transitions in $Ag_{1-x}In_x$ alloys

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The dielectric functions of disordered $Ag_{1-x}In_x$ alloys ($x \leq 0.12$) were obtained ellipsometrically in the 1.2–5.5-eV energy region. In the low-energy region, the intraband scattering rate, described by $\tau^{-1} = \tau_0^{-1} + \beta\omega^2$, increases with increasing In concentration. The increasing positive values of $\beta$ with increasing In content do not agree with a calculation based on the assumption of a spherical Fermi surface and isotropic scattering from an impurity represented by a screened Coulomb potential. The onset energy of the $L_3 \rightarrow L_2'(E_F)$ transition, 4.03 eV for pure Ag, shifts to higher energies, while that of the $L_2'(E_F) \rightarrow L_1$ transition, 3.87 eV for pure Ag, shifts to lower energies. This is only partly attributable to the rise of the Fermi level $E_F$ caused by an increase in the average number of electrons per atom due to the In solute and to the narrowing of the Ag 4$d$ bands. The $L_1$ band may also lower as In is added.

## INTRODUCTION

The optical properties of Ag and Ag-based alloys have been extensively studied to obtain information on the electronic structure of Ag and how it is altered by alloying.[1–11] At low photon energies, below the interband absorption edge, the optical properties of Ag can be explained by the Drude model if a frequency-dependent intraband relaxation rate $\tau^{-1}$ is assumed. Changes in the Drude parameters are expected to occur upon alloying, but the understanding of this effect is still very limited. At higher photon energies, a strong interband absorption edge in $\epsilon_2$ is found at about 3.9 eV and it is known to have a composite nature, the strong $L_3 \rightarrow L_2'(E_F)$ transitions ($d$ band to Fermi surface near point $L$) being overlapped by weak $L_2'(E_F) \rightarrow L_1$ transitions (Fermi surface near point $L$ to a higher conduction band).[2–4] Adding In to Ag causes the $L_3 \rightarrow L_2'(E_F)$ transition to shift to higher energies and the $L_2'(E_F) \rightarrow L_1$ transition to lower energies. However, there have been disagreements among authors about the transition energies[1,4–6] and the shift rates[6,7] upon alloying with In of the two adjacent transition edges.

It is well known that studies of the optical properties of metals and calculations of the one-electron band structures of metals complement each other. The optical constants cannot be interpreted in any detail without some knowledge of the band structure, while band-structure calculations can be checked, and parameters adjusted, by comparison with observed optical spectra.

In the earlier studies of the optical properties of metal alloys the rigid-band model[12] was used to predict the shift of absorption edges by assuming that the alloy has the same band structure and density of states as the host and that the perturbation in the crystal potential caused by the solute simply shifts the energy eigenvalues of the $sp$ and $d$ bands of the host band structure by constant amounts. Thus, the band structure shifts rigidly in energy and $E_F$ shifts with respect to the bottom of the conduction band by an amount determined by the density of states of the hose when the solute valence differs from that of the host. However, many subsequent measurements cast doubt on the applicability of the rigid-band model because this model frequently overestimates the shift of $E_F$.

Another model for estimating the shift of $E_F$ was proposed by Friedel[13] which describes the screening of the excess charges on the solute atoms by the conduction electrons. It leads to very small changes in $E_F$ with respect to the vacuum level in dilute alloys.[14] The host bands lower, especially the $s$-like parts, to accommodate the additional solute electrons. This screening model usually underestimates the shift of $E_F$ upon alloying.[15] Both theories have in common their attention mainly on the displacement of $E_F$. The actual magnitude of shifts in transition energies to or from the Fermi surface can serve as a check on the validity of the two models on dilute alloys. It also aids in correlating optical transitions with band-structure calculations.

There have been theoretical calculations for disordered Cu- and Ag-based alloys (e.g., Cu-Zn, Ag-Cd) within the framework of the average–$t$-matrix approximation (ATA) (Refs. 16 and 17) and the coherent-potential approximation (CPA) (Refs. 18–23) which compute complex bands with real and imaginary parts corresponding to the quasiparticle energy and lifetime. Since such systems lack the translational symmetry of ordered crystals, their eigenstates cannot be characterized in terms of the energy-band picture associated with Bloch's theorem. In the Cu-Zn alloy system where the 3$d$-band complex of Cu is widely separated from that of Zn (by approximately 5 eV) (Ref. 24) the two 3$d$-band complexes are essentially independent and the hybridization between the 3$d$ bands of Zn and the $sp$ bands of Cu is negligible. These models also show the drop in the host bands with respect to $E_F$ as the impurity is added and a narrowing of the $d$ bands as the host $d$ electrons overlap less by dilution. Thus in these models, many changes in the band structure occur. Ag-Cd has been treated by this model.[17]

In the following, we report ellipsometric measurements on pure Ag and $Ag_{1-x}In_x$ alloys ($x=0.04$, 0.08, 0.12) in the 1.2–5.5-eV region to study the evolution of the optical properties related to the intraband and interband transitions in Ag. The Ag-In alloy system has an advantage that the center of the 4*d*-band complex of pure Ag is very widely separated (by approximately 10 eV) (Ref. 24) from that of In so that the hybridization between the 4*d* bands of In and the *sp* bands of Ag is negligible and the effect caused by changing the composition can be more easily resolved. Here, we used the composition-modulation mechanism[3,25] of $\epsilon_2$ in analyzing the data in the higher-energy region. By using a modulation method, sharp structure is obtained in spectra of $\Delta\epsilon_2$ $[=\epsilon_2(\text{alloy})-\epsilon_2(\text{pure})]$ which evolves as In is added. The evolution is attributable to the changes in the band structure of Ag.

## EXPERIMENT

A detailed description of the scanning photometric ellipsometric with rotating polarizer and analyzer used in this measurement was given in Ref. 26. The light sources were a 150-W Xe arc lamp and a 100-W quartz halogen filament lamp with photon-energy ranges of 1.5–5.5 and 1–3 eV, respectively, and a 0.25-m monochromator was used. As detectors photomultipliers with an S20 cathode with an operational energy range of 1.3–6 eV and S1 cathode with 1–3-eV range were used. The amplitudes of the ac output signal from the photomultiplier at $\Omega$, $2\Omega$, and $3\Omega$, with $\Omega/2$, and $\Omega$ ($f=51$ Hz) the angular frequencies of the rotating polarizer and analyzer, respectively, were used to calculate $\tilde\rho$ $(=\tilde r_p/\tilde r_s)$, the complex reflectance ratio between the $p$ (=parallel) and $s$ (=perpendicular) field components of the light beam defined with respect to the plane of incidence of the sample. For an ideal two-phase situation where the reflecting system consists only of an optically thick sample and a transparent ambient with $\epsilon=1$, the complex dielectric function $\tilde\epsilon$ of the sample is related to $\tilde\rho$ through the equation

$$\tilde\epsilon=\sin^2\phi+(\sin^2\phi)(\tan^2\phi)[(1-\tilde\rho)/(1+\tilde\rho)]^2 , \tag{1}$$

where $\phi$ is the angle of incidence of the light beam, which is 68° in this experiment.

Samples were polycrystals made in Ames Laboratory. They were mechanically polished with abrasives, the final grade being a past of 0.05-$\mu$m-diam alumina, and then cleaned with methanol. The measurements were carried out within a few minutes after the samples were cleaned.

## RESULTS

In Fig. 1 the $\epsilon_2$ spectra of various $Ag_{1-x}In_x$ alloys are presented. As In is added we can clearly see the splitting of the two transition edges, the strong $L_3\rightarrow L_2'(E_F)$ around 4.1 eV and the weak $L_2'(E_F)\rightarrow L_1$ around 3.8 eV, which were not resolved in pure Ag.[3-5,9,10]

We can also see the increase in the magnitude of $\epsilon_2$ at low photon energies as the In concentration increases. This can be interpreted in terms of the increased intraband scattering rate of the conduction electrons below the interband absorption edge due to the added impurities.

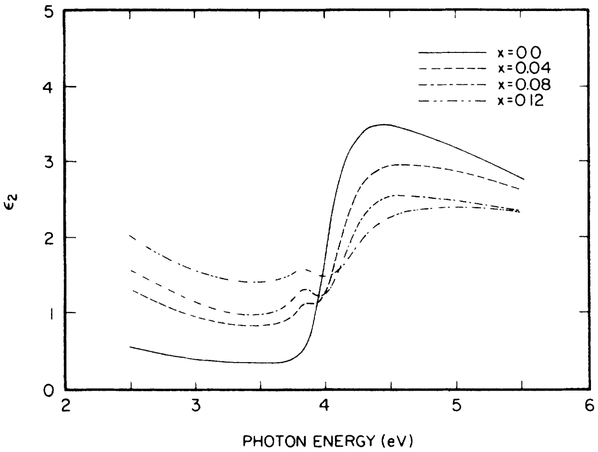

FIG. 1. Imaginary part of the complex dielectric function $\epsilon_2$ vs photon energy for $Ag_{1-x}In_x$.

The low-energy region of the spectra of noble metals arises solely from intraband (Drude) absorption. Below the onset of the first interband transition, around 3.9 eV in Ag, the dielectric function in the simplest Drude model is

$$\begin{gathered}\epsilon_1=\epsilon_\infty-(\omega_p\tau)^2/(1+\omega^2\tau^2) ,\\ \epsilon_2=\omega_p^2\tau/[\omega(1+\omega^2\tau^2)] .\end{gathered} \tag{2}$$

$\omega_p$ is the plasmon frequency, $\tau$ a relaxation time, and $\epsilon_\infty$ the low-frequency dielectric constant due to interband transitions. All three are constants. Experimental data do not fit these equations well. Near the interband threshold, $\epsilon_\infty$ exhibits the dispersion expected from the nearby absorption. More significantly, $\tau$ is not a constant even away from this threshold. $\tau$ is given by

$$\tau^{-1}=\tau_0^{-1}+\beta\omega^2 , \tag{3}$$

which is best viewed as an empirical relation. Causality requires that $\omega_p$ also be frequency dependent,[27] but this dependence has not been observed. For noble metals, the origins of the $\beta$ term are electron-electron scattering,[28-31] electron-phonon scattering,[30,32] and electron-impurity scattering,[32] where applicable. Quantitative agreement between calculated values of $\beta$ and experiment have not yet been achieved.[32] Tables I shows the values of the Drude parameters obtained from our measurements.

In Fig. 2 the volume energy-loss function, $\text{Im}(-1/\tilde\epsilon)$, which is proportional to the probability that an energetic electron will lose energy $E$ by producing excitations in the volume of the system, is presented. In the case of pure Ag, the energy of the first interband transition around 3.9 eV and its strength are within the range of values needed to shift the zero of $\epsilon_1$ from around 9.2 eV to around 3.8 eV. The collective motion of the electrons is now the result of their mutual Coulomb repulsion screened by the polarization of the $d$ band electrons responsible for the interband transition, and an implicit equation for the frequency of the screened plasma oscilla-

TABLE I. Least-squares fitted values of free-electron Drude parameters at low photon energies ($E<2.5$ eV) for $Ag_{1-x}In_x$.

| $x$ | $\epsilon_\infty$ | $\omega_p$ ($10^{16}$ $s^{-1}$) | $1/\tau_0$ ($10^{14}$ $s^{-1}$) | $\beta$ ($10^{14}$ $s^{-1}eV^{-2}$) |
|---|---|---|---|---|
| 0.0 | 2.98 | 1.39 | 0.54 | 0.09 |
| 0.04 | 2.33 | 1.20 | 1.26 | 0.58 |
| 0.08 | 1.40 | 1.14 | 1.92 | 0.78 |
| 0.08 (film) | 1.69 | 1.06 | 1.94 | 0.64 |
| 0.12 | 0.81 | 1.05 | 3.15 | 1.89 |

tions $\omega_p'$ is

$$\omega_p'=\omega_p/[1+\epsilon_1^b(\omega_p')]^{1/2}\ , \tag{4}$$

where $\omega_p'\tau>>1$, $\epsilon_2^b(\omega_p')\simeq 0$, and $\epsilon_1^b$ and $\epsilon_2^b$ are the interband terms of $\tilde{\epsilon}$. Equation (4) is valid for pure Ag, but not for the alloys we have studied. For them, $\epsilon_2^b$ is not negligible, as we can see in Fig. 1, so that Eq. (4) must be generalized.

For pure Ag, $\mathrm{Im}(-1/\tilde{\epsilon})$ shows a sharp peak at 3.77 eV in our experimental data, as can be seen in Fig. 2, and as In is added the broadening and splitting of the structure into two small peaks are seen, which is indirect evidence of the splitting of the two transition edges. Figure 3 shows $\Delta\epsilon_2$ $(=\epsilon_{2(\mathrm{alloy})}-\epsilon_{2(\mathrm{pure})})$ spectra. We see two peaks, positive and negative, due to the shifts of the two transition edges, $L_2'(E_F)\rightarrow L_1$ and $L_3\rightarrow L_2'(E_F)$, respectively, and the position of the positive peaks shifts to lower energies while that of the negative peak shifts to higher energies. The positions of the two peaks are listed in Table II and they nearly coincide with the maximum of the numerically calculated first derivative of $\epsilon_2$ within 0.02 eV, the interval of scanned photon energies. For pure Ag, we take the photon energy leading to the minimum reflectivity as the $L_2'(E_F)\rightarrow L_1$ transition edge and take the photon energy leading to the maximum of the numerically calculated $d\epsilon_2/d\omega$ as the $L_3\rightarrow L_2'(E_F)$ transition edge. Their energies were found to be 4.03 and 3.87 eV which agree well with the result in Ref. 4. We can estimate the shifts of the two transitions edges due to alloying by just subtracting the transition energies for pure Ag from the peak energies in Fig. 3.

## DISCUSSION

Figure 4 shows the dependence of the Drude parameters on the concentration of In. For pure Ag, the value of $\tau_0^{-1}$ from the optical data is about 20% of the value obtained from the dc resistivity. $\tau_0^{-1}$ increases linearly with increasing In concentration, but at a rate only about one-third the rate of increase of the dc resistivity.[33] $\beta$ similarly increases approximately linearly with In concentration. This is surprising in view of the one calculation of the effect of impurities on $\beta$.[32] That calculation assumed a spherical Fermi surface and isotropic scattering from an impurity represented by a screened Coulomb potential. It prediced a $\beta$ that was linearly proportional to the impurity concentration, but with a negative sign for all reasonable values of the screening length. With this model, a positive $\beta$ results only if the screening length becomes less than a few tenths of an angstrom. The positive values of $\beta$ we have obtained indicate that this model needs to be extended. It is known that the screened Coulomb potential predicts a value for the rate of change of the dc resistivity with impurity concentra-

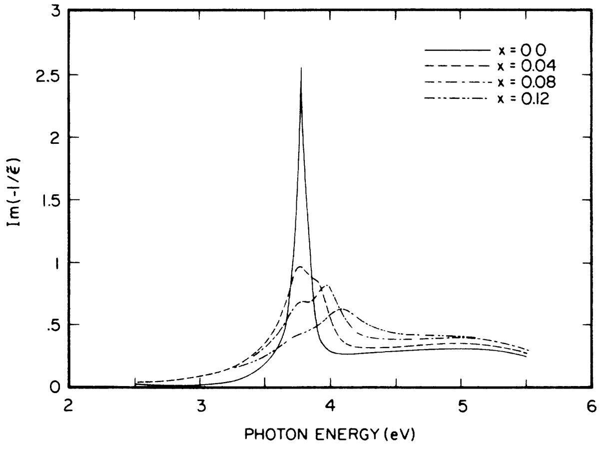

FIG. 2. Volume energy-loss function vs photon energy for $Ag_{1-x}In_x$.

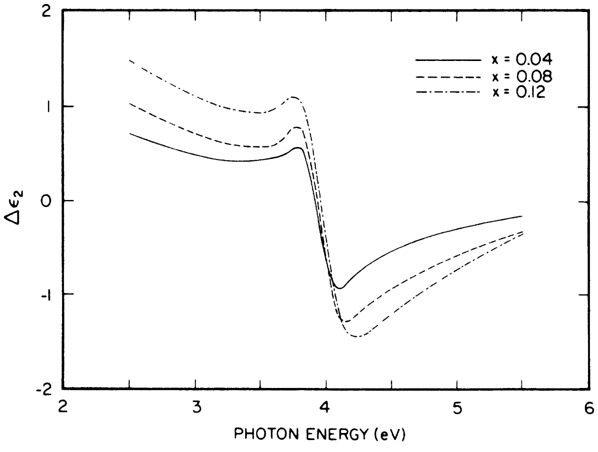

FIG. 3. The change in the imaginary part of the complex dielectric function $\Delta\epsilon_2$ vs photon energy for $Ag_{1-x}In_x$.

TABLE II. Photon energies of the two transition edges obtained by taking the two (negative and positive) peak positions in the $\Delta\epsilon_2$ spectra for $Ag_{1-x}In_x$.

| $x$ | $L_3 \rightarrow L_2'(E_F)$ (eV) | $L_2'(E_F) \rightarrow L_1$ (eV) |
|---|---|---|
| 0.0 | 4.03[a] | 3.87[b] |
| 0.04 | 4.11 | 3.81 |
| 0.08 | 4.17 | 3.79 |
| 0.12 | 4.23 | 3.77 |

[a] Photon energy of maximum $d\epsilon_2/d\omega$ for pure Ag.

[b] Photon energy of minimum $R$ (reflectivity) for pure Ag.

TABLE III. Comparison of the experimentally determined Fermi-level shifts $\Delta E_F$ with calculations based on the Friedel model and the rigid-band mode for $Ag_{1-x}In_x$.

| $x$ | This work (eV) | Friedel (eV) | Rigid band (eV) |
|---|---|---|---|
| 0.04 | 0.06 | 0.01 | 0.29 |
| 0.08 | 0.08 | 0.02 | 0.58 |
| 0.12 | 0.10 | 0.04 | 0.88 |

tion that is too high,[34] one that is improved by using the Friedel model.[35] Moreover, impurity (Sn,Au) scattering on the Fermi surface of Ag is known to be anisotropic,[36] so a nonspherical Fermi surface may also have to be considered. Anisotropic scattering by a vacancy in Cu and Au has been calculated recently.[37]

The simplest model for alloys is the rigid-band model. According to this, the Fermi level will rise as the addition of In increases the electron-to-atom ratio. Using the electronic specific heat[9,38] as a measure of the density of states at the Fermi level, this rise is 0.29 eV for 4% In (Table III). The $L_3 \rightarrow L_2'(E_F)$ transition should then increase by this amount, and the $L_2'(E_F) \rightarrow L_1$ transition decrease by nearly 0.29 eV, the band along $Q$ near $L_1$ being quite flat. This gives the right sign for the shifts, but grossly overestimates their magnitudes.

The Friedel model emphasizes the small change in the Fermi level as the polyvalent solute is added, but this is with respect to its original value. The extra electrons are accomodated below $E_F$ by the downward motion of the bands, especially the $s$ bands, whose electrons best sample the unscreened center of the impurity potential. In the band picture this can be mimicked to zero order by leaving the Fermi level fixed and lowering the conduction bands, the 4$d$ bands remaining unshifted because of the better localization of the 4$d$ electrons. The end result is that the $L_2' \rightarrow L_1$ transition energy should decrease by more than 0.29 eV, the additional shift being due to the downward shift of $L_1$ ($s$-like) with respect to $L_2'$ ($p$-like), and the $L_3 \rightarrow L_2'$ transition energy should increase by the amount the Freidel screening model predicts the Fermi energy to rise, for this rise may be measured against the fixed $d$ band. For 4% In, this is about 0.01 eV as listed in Table III.

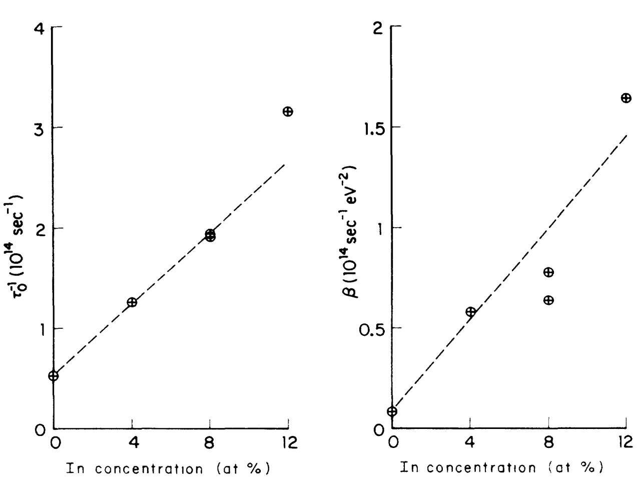

FIG. 4. The dependence of the Drude parameters on In concentration.

That such an increase of the Fermi level with respect to the bands actually occurs is borne out by the measured increase of the neck of the Fermi surface of Ag as Mg or Cd are added,[39] and of Cu as diverse polyvalent solutes are added.[40,41]

An additional effect expected is that as the solute is added—any solute—the $d$ bands narrow because of the missing $d$-$d$ overlap as nearest-neighbor solvent atoms are replaced by solute atoms. This lowers $L_3$ and further increases the energy of the $L_3 \rightarrow L_2'(E_F)$ transition, already too large from the Fermi-level motion in the rigid-band model, but in a direction to improve agreement with the Freidel model. The shift of the top of the 4$d$ band as In is added can be seen in photoemission measurements.[42,43] There the 4$d$ band narrows by 0.132 eV at 4% In.[43] If the band narrows symmetrically, the $L_3$ point then drops 0.066 eV below $E_F$. In fact, the top of the 4$d$ band should drop more than the bottom of the 4$d$ band rises.

The above-mentioned picture can be applied to the corresponding transitions in Cu as polyvalent solutes are added. Cu is a solvent whose band structure is better understood than that of Ag, and a wider variety of solutes have been studied. The rise in the Fermi energy for 4% Ga, the 3$d$ analog of In, is 0.27 eV from the electronic specific heat.[44] The observed shifts of the $L_3 \rightarrow L_2'(E_F)$ and $L_2'(E_F) \rightarrow L_1$ transition energies are $+0.054$ and $-0.52$ eV, respectively, from Staines's piezoreflectance study of Cu alloys.[45] Note that the shift of the $L_2' \rightarrow L_1$ transition is about a factor of 8–9 larger in Cu-Ga than in Ag-In. The $L_3 \rightarrow L_2'(E_F)$ shift can be reconciled with the Freidel model if the 3$d$ band narrows, but the $L_2'(E_F) \rightarrow L_1$ shift is too large. Staines[45] and others[46,47] analyzed these results further, but there is not still good agreement between experiment and theory. An example is that the $d$ bands should narrow for increasing solute addition, no matter what the solute. Pells and Montgomery[48] showed that for Cu alloys the narrowing was more nearly constant for equal electron-to-atom ratios rather than equal impurity concentrations.

Modern alloy theory allows a better understanding. CPA calculations incorporate all of the above-mentioned effects. Adding polyvalent solutes to Cu raises the Fermi level some, lowers the conduction bands, especially the *s*-like parts, and narrows the *d* bands. Calculations have been carried out for several such solutes in Cu,[16,22,23,46] but not all the solutes for which data exist, and qualitative agreement for most of the shifts exist. For these alloys, the $L_2'$-$L_1$ gap was adjusted[23] to fit optical data. All other calculated gaps were then in agreement with experiment. For Ag alloys, only two calculations exist, an empirical calculation,[49] fitting parameters to experimental data, and one using the average–*t*-matrix approximation.[17] Both were for Ag-Cd, and both began with parameters for the bands of Ag that do not give results in as good agreement with a variety of experimental data as do the corresponding parameters for Cu. They found that, with respect to a fixed Fermi energy, the *d* bands dropped some, and the *sp* bands dropped even more, as Cd was added. In addition, the empirical calculation required the $L_2'$-$L_1$ gap to decrease significantly in order to produce agreement with experiment.

The $L_2'$-$L_1$ gap is very sensitive to a number of parameters. It is volume sensitive. However, the fractional volume increases upon adding In to Ag and Ga to Cu are nearly identical.[50] The calculated deformation potentials for this gap, although they differ by about a factor of 2 in the two calculations done to date,[51,52] are nearly the same for Cu and Ag.[52] [The smaller of the two values for Cu (Ref. 51) leads to a shift of the $L_2'(E_F)\rightarrow L_1$ transition that is about half of that observed.[53]] This gap is also sensitive to shifts in the $L_3$ level, for the $L_1$ level interacts strongly with the $L_3$ level, via orthogonalization and second-order hybridization.[54] Finally, the $L_1$ level is sensitive to the impurity potential more than the $L_3$ and $L_2'$ levels, for the *s* electrons sample the impurity core. All these features are taken into account in some approximation in CPA or average–*t*-matrix calculations, with the former (not carried out yet on Ag alloys) treating the possible charge transfer that the latter does not treat. The shift of the $L_2'$-$L_1$ gap upon alloying is important in understanding the optical properties of Cu-, Ag-, and presumably, Au-based alloys with elements of larger valence. This shift is significantly greater for Cu-Ga than for Ag-In, for reasons that are not completely clear at present.

## SUMMARY

The addition of In to Ag causes the Drude parameters to change. The frequency dependence changes with a sign not in accord with a calculation based on a relatively simple model of impurity scattering. The interband absorption shifts cannot be explained only by an increase in the Fermi level and a narrowing of the 4*d* band. A decrease in the $L_2'$-$L_1$ gap is necessary. Comparisons with Ga in Cu show similar effects, but the shifts are larger than in Ag with In.

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