Optical absorption of *Ni*Cu alloys

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A study of the optical absorption of *Ni*Cu alloys for Cu concentrations from 2 to 13 at.% by a sensitive differential technique is presented. The experiment is analyzed for the difference in the optical conductivity between the alloy and pure Ni. The results are discussed in terms of the generalized Anderson model of dilute alloys. We also discuss the possible presence of k-non-conserving optical-absorption processes induced by the Cu impurities. The data appear to be consistent with the coherent-potential-approximation calculations of the density of states for *Ni*Cu alloys. Edges at $\hbar\omega = 0.9$ eV and $\hbar\omega = 1.7$ eV are interpreted in terms of impurity density-of-states peaks 0.8 and 1.7 eV below E_F. The shape of the structure between 1.7 and 3 eV is consistent with an exchange splitting of about 0.6 eV in ferromagnetic Ni.

I. INTRODUCTION

The best understood disordered transition-metal alloys are those with noble-metal hosts and low concentrations of Ni or Pd.[1,2] In these alloys the impurity d levels fall above the host d bands, and lie in the nearly-free-electron s-p bands with which they weakly mix to form virtually bound states. The optical properties of these alloys are particularly simple as the absorption associated with the impurity d levels falls at lower frequencies than the interband edge, and is therefore superimposed on a smooth structureless background. Optical studies of *Cu*Ni,[3-5] *Au*Ni,[4,6] *Cu*Pd,[7] *Ag*Pd,[7-11] and *Au*Pd[7] have proven very fruitful. Also, photoemission studies on these alloys display the impurity density of states clearly separated from the host d bands.[12]

More generally the impurity d levels fall within the host d bands in transition-metal alloys.[1] In this case the impurity density of states is expected to have a larger energy width and a more complex spectrum. The theory of the electronic structure of these alloys is much more complex so that our understanding of them is much poorer. Unfortunately the experiments are also more difficult to interpret in this case since the host d bands also change upon alloying, and it is difficult to separate these changes from those due to the added impurity levels (assuming such a separation is justified). Consequently there have been few measurements and still fewer interpretations of optical or photoemission spectra for these alloys, even though these are just the alloys of the most technological interest.

In this paper we present a study of the optical properties of *Ni*Cu alloys.[13] The Cu-Ni alloy series has been widely studied both experimentally and theoretically. In the Cu-rich alloys, the Ni-impurity d levels form a virtually bound state, and this system is reasonably well understood.[2,3] We are interested in attempting to interpret the optical spectra of the Ni-rich alloys in terms of the modern theory of transition-metal alloys. One feature of these alloys that makes it feasible to interpret their optical-absorption spectra is that the final states of the absorption process are fairly simple. Since the d bands lie almost entirely below E_F with only the minority spin d band extending a small fraction of an eV above the Fermi level, the final states in the absorption process lie either in the flat s-p bands or in the very sharp Ni d band at the Fermi level.

The paper is organized as follows. The experiment is described in Sec. II and the interpretation of the optical data is discussed in Sec. III. Concluding remarks are made in Sec. IV.

II. EXPERIMENTAL

The samples were prepared by simultaneous vacuum evaporation of the two constituents onto polished fused quartz substrates, 1 mm thick by 25.4 mm diam. Two substrates were mounted in the evaporation chamber in the geometry shown in Fig. 1, so that a shield placed between the two sources prevented the copper vapor from falling onto the pure substrate while permitting the nickel

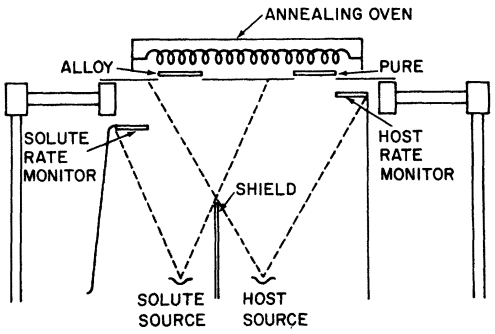

FIG. 1. Configuration of the vacuum evaporator used for the simultaneous production of pure metal and alloy films.

vapor to fall on both. The deposition rates were typically 50 Å/sec (monitored by a quartz oscillator), and the total thickness was typically 3000 Å. The nickel was evaporated with a Varian 2-kW electron-beam evaporation source, while copper was evaporated from a tungsten boat by resistive heating. The evaporation chamber was evacuated by a conventional oil diffusion pump with liquid-nitrogen cold trap. During film deposition the pressure was about 5×10^{-6} Torr and during the subsequent annealing (400 °C for 15 min) it went down to about 5×10^{-7} Torr. A longer anneal resulted in clouding of the pure-nickel sample, indicating that recrystallization had reached the scale of the wavelength of light. Lower annealing temperatures were also unsatisfactory because the reflectivity results were then found to change for several days, as the samples continued to anneal at room temperature. The 400 °C annealing temperature produced stable results. The copper concentrations were determined from dc resistivity ratios of the samples between room temperature and 4 °K using the resistivity data of Svensson.[14] Corrections due to surface scattering were made empirically based on chemical analysis of several of our films. Our concentration determination was accurate to about 10% relative.

Reflectivity measurements were made with a single beam differential reflectometer similar to that described by Beaglehole.[15] Both pure and alloy samples were mounted on a rotating holder so that light was reflected alternately from a pure-nickel and an alloy sample. The quantity $\alpha\equiv(R_{\text{pure}}-R_{\text{alloy}})/(R_{\text{pure}}+R_{\text{alloy}})$ was recorded continuously as a function of photon wavelength.

A typical spectrum of α vs ω is shown in Fig. 2. α was measured to one part in 10^4 in the visible, and the precision fell off to one part in 10^3 in the infrared (below 0.6 eV) and in the ultraviolet (above 5 eV). Only pure and alloy samples prepared at the same time were compared with one another in order to reduce the reflectivity differences due to variations of vacuum evaporation conditions and the effects of "aging" after exposure to air. The reflectivities of our pure-nickel films were measured separately using a Perkin-Elmer model 350 spectrophotometer with a specular reflectance accessory. We found agreement to within a few percent over the measured spectral range with the published results on bulk samples.[16-18]

The complex conductivity function $\sigma=\sigma_1+i\sigma_2$ of the sample was obtained through a Kramers-Kronig analysis of the reflectivity data. σ_{pure} and σ_{alloy} were determined separately from R_{pure} and $R_{\text{alloy}}=R_{\text{pure}}[(1-\alpha)/(1+\alpha)]$. The results were then presented in terms of the differential conductivity $\Delta\sigma_1=\sigma_1^{\text{alloy}}-\sigma_1^{\text{pure}}$. In order to perform the Kramers-Kronig analysis it was necessary to extrapolate the reflectivity data outside the measured spectral range. For the low-energy extrapolation we modeled the complex dielectric function $\epsilon=1+4\pi i\sigma/\omega$ as the sum of a Drude term for the conduction-electron response and a simple Lorentzian oscillator term to account in an approximate way for the interband absorption. The oscillator frequency was located well above the low-frequency cutoff, ω_l, at 0.5 eV. Thus for the low-energy extrapolation, we have

$$\epsilon(\omega)=1-\frac{\omega_p^2}{\omega(\omega+i/\tau)}+\frac{f}{\omega_0^2-\omega^2-i\Gamma\omega},\quad \omega<\omega_l. \tag{1}$$

For the Drude term we used the plasma frequency ($\omega_p=8.3$ eV) given by Ehrenreich *et al.*[19] for pure nickel, and assumed that it was unchanged for the

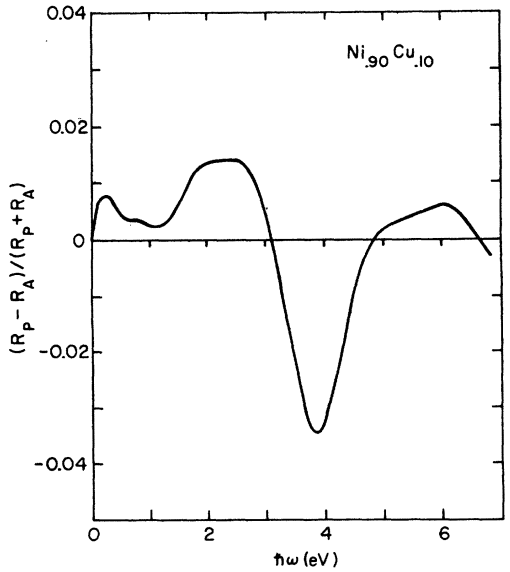

FIG. 2. Typical spectrum of $\alpha=(R_P-R_A)/(R_P+R_A)$ measured for a *Ni*Cu alloy film.

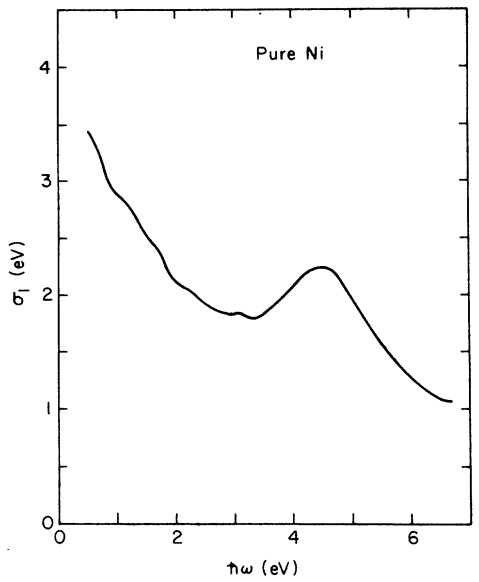

FIG. 3. Optical conductivity of pure nickel as obtained from a Kramers-Kronig analysis of reflectivity data as described in the text.

alloy (for pure Cu $\omega_p = 9.3$ eV). The lifetime τ was taken as the zero-frequency value determined from the dc resistivity. The three parameters in the Lorentzian oscillator term were then determined by a least-squares fit to the reflectivity data near the low-frequency cutoff at $\hbar\omega_l = 0.5$ eV. This procedure is found to provide a physically consistent low-energy extrapolation. However, the results for $\Delta\sigma$ are not very sensitive to this extrapolation, even if much simpler procedures are used, except very near the cutoff frequency.

We extrapolated our reflectivity data for pure Ni to higher energy ($\omega > 6.8$ eV) so that the Kramers-Kronig inverted optical conductivity, $\sigma_1(\omega) = \omega\epsilon_2(\omega)/4\pi$, matched the ellipsometric data of Kirrilova[18] around 2 eV where ellipsometry is considered most reliable. $\sigma_1(\omega)$ obtained in this manner for our pure-Ni film is shown in Fig. 3. For alloy samples we modified the extrapolation in order to satisfy the differential conductivity sum rule

$$\Delta n = \frac{2m}{\pi e^2}\int_0^{\omega_s} \Delta\sigma_1(\omega')\,d\omega', \qquad (2)$$

where Δn is the difference of the density of valence electrons between pure Ni and the alloys. The high-frequency cutoff ω_s in Eq. (2) was taken to be the frequency for the saturation of the sum rule for valence electrons in the pure system. We chose ω_s to be 100 eV, corresponding to the experimental result for pure Cu.[20] Although the magnitude of $\sigma_1(\omega)$ within the data range is in general strongly dependent on the choice of the high-energy extrapolation function, the results for $\Delta\sigma_1 = \sigma_1^{\text{alloy}} - \sigma_1^{\text{pure}}$ obtained this way were practically insensitive to the extrapolation functions.

III. DISCUSSION

The results for $\Delta\sigma$ for five *Ni*Cu compositions between 2- and 13-at.% Cu are shown in Fig. 4. We will be discussing the following spectral features in this section. The low-frequency part (below 1 eV) contains a sharply rising low-frequency divergence and a shoulder around 0.9 eV. The second region of interest is from 1 to 3 eV. One sees an edge at about 1.7 eV, a shoulder at 1.9 eV, and a peak around 3 eV. Finally, at high frequencies, there is structure that appears to be related to the 4.5-eV peak in the conductivity of

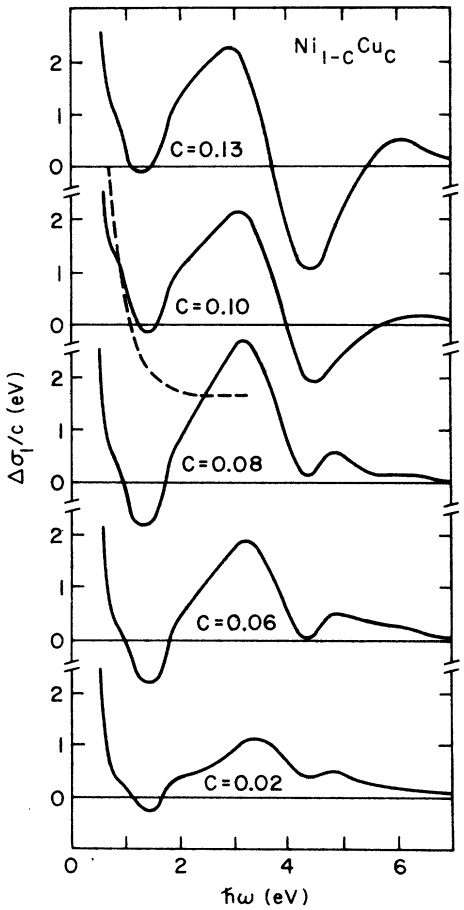

FIG. 4. Measurements of the differential optical conductivity per atomic percent for a series of *Ni*Cu alloys. The dashed curve represents a model $\Delta\sigma/c$ consisting of a Drude contribution estimated from the dc conductivity and a uniform reduction of the host conductivity for the the case of a 10% alloy.

pure Ni. The structure in this range appears more nonlinear with Cu concentration than the rest of the spectrum. The smaller $\Delta\sigma/c$ for the 2% alloy may result from an inaccuracy in the determination of the Cu concentration for this alloy.

We will discuss the interpretation of the $\Delta\sigma$ spectra in terms of three processes: (a) Modifications in the host density of states which can include a reduction due to the replacement of host atoms with the impurity atoms and shifts and broadening of host bands. (b) Absorption associated with the impurity density of states. This includes transitions from impurity states to host states and the modification of the intraband absorption due to the impurity. (c) Impurity-induced k-nonconserving interband absorption. In this process it is expected that optical edges can occur at frequencies corresponding to the separation of peaks in the host density of states and the Fermi level, and optical-absorption peaks can occur at frequencies corresponding to the separation of occupied and unoccupied peaks in the host density of states.

Of these three processes, (a) and (b) have been considered by Beaglehole and Hendrickson[21] in analyzing and interpreting their reflectivity data from dilute *Au*Fe alloys. From a theoretical standpoint, only process (b) has been usefully developed in the literature.[22] Several authors have considered the optical response within the Anderson model of dilute alloys.[22-24] They have only considered the case where the impurity is placed in a simple free-electron-like band. Since the final states in our experiment would be the free-electron-like s-p bands about E_F, these calculations are applicable.

The result for $\Delta\sigma/c$ in terms of the impurity density of states, $\rho_s^I(E)$, is

$$\Delta\sigma = \sum_s \frac{c}{8\omega}\left(\frac{\omega_p^2 V^2}{\omega^2} + \omega_d^2\right)\int_{-\omega}^{0} dE\,\rho_s^I(E)\,, \tag{3}$$

where c is the impurity concentration and s is the electron spin. ω_p is the effective plasma frequency associated with the s-p band and is 8.3 eV for Ni. ω_d is related to the d-s matrix element for the impurity and it can be estimated from the strength of the interband edge in pure Cu to be of order 2–3 eV.

We will first ignore process (c) and interpret our data in terms of a model including only processes (a) and (b). We will consider later how our interpretation could be modified if process (c) is included.

The low-frequency divergence in $\Delta\sigma$ can be interpreted largely in terms of the increased scattering of the conduction electrons in the alloy. The contribution of the Drude term to the conductivity is

$$\sigma = \sigma_0/(1 - i\omega\tau)\,, \tag{4}$$

where $\sigma_0 = ne^2\tau/m^*$. At the frequencies we are dealing with $\omega\tau \gg 1$ and so we can approximate (for the sake of discussion)

$$\mathrm{Re}\sigma \simeq \frac{\sigma_0}{(\omega\tau)^2} = \frac{1}{4\pi}\frac{\omega_p^2}{\omega^2}\frac{1}{\tau}\,. \tag{5}$$

Therefore $\Delta\sigma \simeq (1/4\pi)(\omega_p^2/\omega^2)\Delta(1/\tau)$. Equation (5) clearly gives us a low-frequency divergence in $\Delta\sigma$ as observed experimentally. However, we should subtract from this a term $c(\sigma_P - \sigma_D)$, where σ_P stands for σ_{pure}, and σ_D is the Drude part of the conductivity of the pure sample. When this subtraction is performed from $\Delta\sigma$ calculated from Eq. (5) using $\Delta(1/\tau)$ estimated from the dc resistivity, we get the result illustrated in Fig. 4 as a dashed line. Limitations of the data and the Kramers-Kronig analysis make it pointless to elaborate on this discussion. However, the result does adequately describe the low-frequency behavior of $\Delta\sigma$.

The optical conductivity of pure Ni (Fig. 3) is smooth compared with our $\Delta\sigma$ spectra (Fig. 4). The most notable structure in σ_{pure} is the peak at 4.5 eV. Therefore we expect a negative contribution to $\Delta\sigma$ as discussed in (a) that should be smooth except near 4.5 eV. In the simplest model this contribution would go like $\Delta\sigma \sim -c\sigma_{\text{pure}}$. Therefore, within the model we are considering [ignoring (c) process], the $\Delta\sigma$ structure around 0.9 eV and from 1.7–3 eV would be related to the impurity levels in the alloy.

It is generally expected that the impurity d levels in these alloys are not sharp virtual levels since they fall within the high density of states d bands of the host. Indeed coherent-potential-approximation (CPA) calculations on low Cu concentration paramagnetic *Ni*Cu alloys by Stocks, Williams, and Faulkner[25] indicate a broad Cu density of states centered at about 4 eV below the Fermi level and with a half width of more than 1 eV. Also, we have made calculations based on the Wolff model of dilute alloys[26] using the calculated Ni density of states from Callaway and Wang,[27] and they support this conclusion. Although the impurity density of states is spread over a several eV range, it contains some sharp features which are related to sharp features in the host density of states. For example, the CPA calculation on an 11-at.% Cu paramagnetic alloy shows a peak in $\rho_s^I(E)$ about 2 eV below the Fermi level which contains about 20% of the total Cu states. This peak is associated with a peak in the pure-Ni density of states at about −1.6 eV. It is interesting therefore to consider the possibility that the edge in $\Delta\sigma$ at about 1.7 eV corresponds to transitions from this subsidiary Cu peak. To do this we must first discuss the expected modification of the CPA calculation for the case of ferromagnetic alloys that we

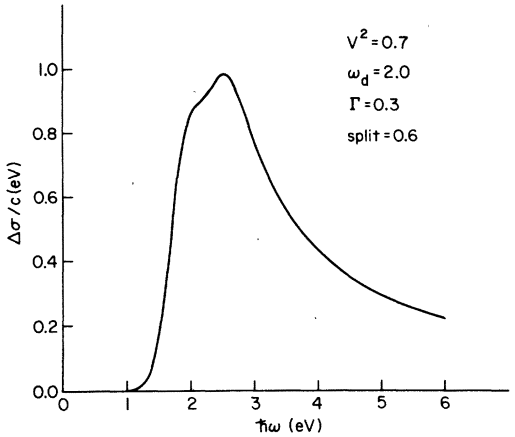

FIG. 5. Calculated contribution to $\Delta\sigma/c$ for a model impurity density of states consisting of two Gaussian peaks at -1.8 and -2.4 eV within the Anderson model. The width parameter of the Gaussians is denoted by Γ.

are dealing with.

Self-consistent band calculations including the exchange interaction by Callaway and Wang[27] indicate that there is a nearly constant exchange splitting Δ between the minority and majority spin d-like bands in ferromagnetic Ni. The density of states for each spin is similar in shape to the paramagnetic case, and the two spins are separated by $\Delta \sim 0.6$ eV.[28] Since the spin-orbit interaction in *Ni*Cu is small and the Cu ion should be nearly nonmagnetic, we expect that the scattering of Bloch electrons from the impurity should not flip spins. Therefore, to a good approximation, we expect the spin-up and spin-down Cu impurity densities of states to be identical in shape and split by Δ. We expect, in other words, two small peaks in the Cu density of states around 2 eV split by the exchange splitting. In Fig. 5 we show a conductivity line shape based on Eq. (3) with two Gaussian peaks located at -1.8 and -2.4 eV, each containing one electron. The half width of the peaks was taken as 0.3 eV. For V^2 we have used the value deduced from the width of the Ni virtual state in *Cu*Ni alloys. This is justified because the V in Eq. (3) is the mixing matrix element between the impurity d state and the final state of the optical-absorption process. The amplitude and structure of this line shape are seen to be in reasonable agreement with the experiments.

What about the position of the edge? In the CPA calculations[25] the Cu peak is at -2.1 eV, about 0.5 eV below the low-energy peak in the Ni density of states in the calculation. (Energy is expressed relative to the Fermi energy.) Callaway and Wang[27] place the lower peak in the pure-Ni minority-spin density of states at -1.7 eV. However, comparison of their calculations of the optical conductivity with optical data indicates that band calculations have gotten the Ni d-band width about 1 eV too wide. Since the Fermi level is fixed near the top of the minority-spin d band, a uniform compression of the d band to correct for the width would shift the -1.7 eV peak to about -1.2 eV. The peak in the Cu impurity state density would then occur at about -1.7 eV, giving rise to an optical edge at $\hbar\omega = 1.7$ eV as is observed.

Can we make a similar interpretation of the $\Delta\sigma$ structure near 0.9 eV? The pure-Ni conductivity is very smooth in this range, supporting the idea that this structure arises from a small peak in the Cu state density at about -0.8 eV. The high-energy peak in the majority-spin band of Ni lies somewhat less than the exchange splitting ($\Delta \sim 0.6$) below E_F so that an impurity structure around -0.8 eV is reasonable. This peak is not seen in the CPA calculations,[25] but it is seen as a small peak in our Wolff-model calculations. We find that we can account for the magnitude of the shoulder in $\Delta\sigma$ at 0.9 eV by assuming a peak in the impurity density of states around 0.8 eV, containing only 0.05 electrons, in Eq. (3).

In the 3–6 eV frequency range the differential conductivity is strongly affected by changes in the large 4.5-eV peak in σ_p. This peak has been identified as most likely resulting from transitions between the lower s-d bands and the s-p bands above the Fermi energy.[27] Shiga and Pells[29] have measured the temperature dependence of the optical conductance of pure Ni, and they find that this 4.5-eV peak narrows as the temperature is raised through the Curie temperature. They interpret the decreasing width in terms of a reduction in the exchange splitting of the s-d bands with increasing temperature.

In our experiment, the structure in $\Delta\sigma$ associated with the 4.5 eV peak in σ is seen to be nonlinear in Cu concentration. There is a negative dip at 4.5 eV which grows more rapidly than linearly with c above 8 at.%.

In the high-frequency range we therefore expect a complicated $\Delta\sigma$ spectrum where at least processes (a) and (b) are both contributing nontrivially. Stocks, Williams, and Faulkner[25] place most of the Cu d states in a broad peak ($\gtrsim 1$ eV half width) centered about 4 eV below the Fermi level. We propose to explain the line shape of $\Delta\sigma$ in this range in terms of our model as follows: at low Cu concentrations we suppose that the two contributions roughly cancel leaving little structure. Comparing $c\sigma_p$ with the impurity contribution calculated from Eq. (3) shows that such a cancellation is not unreasonable. At higher concentrations we suppose that these two contributions no longer

nearly cancel because of nonlinear effects on one or the other or both contributions. One possibility is that the Cu impurity d band begins to broaden when the concentration gets high enough so that the average Cu atom has one or more Cu nearest neighbors. Also, the 4.5-eV structure in the host may narrow because of the reduced exchange splitting in the alloy, and this would tend to influence the position of the spin-up and spin-down Cu impurity levels. Therefore, there are several nonlinear effects occurring simultaneously in this spectral range, and so far we have not been able to make a simple interpretation of the observed nonlinear spectra.

This discussion shows that the measured optical properties of the *Ni*Cu alloys can be reasonably interpreted in terms of a model including only processes (a) and (b) and the current theoretical ideas about the Cu impurity density of states. The arguments on the line shapes of the spectral features are strengthened by estimates of the expected magnitudes of $\Delta\sigma$. If the (c) processes are, in fact, small so that neglecting them was justified, the above interpretation gives the positions and magnitudes of some of the prominent peaks in the impurity density of states for this alloy. It is particularly intriguing to consider whether this experiment provides a measure of the exchange splitting in the alloy. The line shape shown in Fig. 5 was calculated from Eq. (3) including an exchange splitting, Δ, of 0.6 eV. A larger exchange splitting produces a dip above 2 eV, and a smaller value smooths the shoulder at 2 eV excessively. The assumption of double Gaussian density of states peaks is undoubtedly too simple. The peaks are likely to be asymmetrical, and the tail of the remaining state densities at lower energies should contribute in this range (particularly the minority-spin related part). In addition σ_p is not constant in the 1.5–3 eV range so that the (a) process contribution to $\Delta\sigma$ should distort the line shape. Despite these considerations, it seems unlikely that Δ will differ very much from 0.6 eV if we can neglect the (c) process in our interpretation. It is also interesting to note that the peak near 3 eV shifts toward lower energies as the Cu concentration is increased. This is consistent with a reduction of the exchange splitting in the alloy. Indeed the magnitude of the shift is also roughly consistent with the expected shift if it is assumed that Δ is proportional to the magnetization of the alloy. However, we cannot be certain that this shift is not caused by the nonlinear effects occurring in the 4–6 eV range.

How is the interpretation changed if we assume that the (c) processes are not negligible? In this case we must assume that the $\Delta\sigma$ spectrum is a mixture of the three processes at roughly equal magnitudes since we have shown that the magnitudes of processes (a) and (b) are expected to be of the same order as the observed $\Delta\sigma$. If we assume that the line shapes of the $\Delta\sigma$ contribution from the k-nonconserving processes is primarily controlled by the host density of states we can expect the following kinds of structures: (i) Transitions from peaks in the host density of states below E_F to the sharp minority-spin d band at the Fermi level should produce peaks in $\Delta\sigma$. Neglecting spin-flipping transitions, this process should occur only in the minority-spin band since the majority-spin band lies entirely below the Fermi level. (ii) Transition from peaks in the host density of states to s-p bands above the Fermi level should produce optical-absorption edges. In either case, the position of the structure is a measure of the energy of the host density of states feature with respect to the Fermi level.

It is interesting to consider whether the shoulder in $\Delta\sigma$ at 0.9 eV and/or the edge at 1.7 eV and peak at 3 eV may arise from these k-nonconserving transitions. The 1.7-eV edge could come from the lower peak in the minority-spin band which lies about 1.7 eV below the Fermi level according to Callaway and Wang.[27] However, they argue that this peak should be moved to higher energies for two reasons. First, they have gotten the d-band width too great as discussed earlier. Also, they believe that the exchange splitting is too large in their calculation. Therefore, it appears that the k-nonconserving edge from this feature should occur at a frequency somewhat less than 1.7 eV. On the other hand, it would be natural to interpret the shoulder at 0.9 eV in terms of k-nonconserving transition from the large peak at the top of the majority-spin d band in the host. Once again however, the location of the edge is considerably higher than expected. This density of states peak should be nearly Δ below E_F, and Δ is estimated to be about 0.6 eV. We also note that the optical spectra do not appear to contain any structure that has a line shape as expected for case (i) above. However, we recognize that a detailed calculation of these k-nonconserving contributions may not turn out to be so simple as pictured in (i) and (ii) above.

IV. CONCLUSIONS

We have shown that the differential optical conductivity of *Ni*Cu alloys provides a sensitive probe to the electronic structure of the system. However, the data are not susceptible to the kind of line-shape analysis performed on *Cu*Ni and the related alloys because of the combination of several different processes contributing to $\Delta\sigma$.

Rather, it is necessary in this case to make a more qualitative interpretation of the optical data based on band-structure calculations, alloy theory, and estimations of optical conductivity. These arguments are hampered by the lack of an adequate theory of the contribution of k-nonconserving interband transitions in alloys. However, it appears that the interpretation of the data in terms of a contribution due to transitions from impurity levels to band states above the Fermi level, and a negative contribution corresponding to a reduction in the interband conductivity in the alloy, is more consistent with the band-structure calculations for Ni than an interpretation based on the impurity-induced k-nonconserving interband absorption processes. It may be that the contribution of this k-nonconserving transition is small compared with the other processes. Studies of related alloys, such as *Ni*Au and *Ni*Ag and those of Pd and Pt hosts with noble-metal impurities would possibly lead to interesting new results and a clarification of the issues raised by this study. It would also be fruitful to develop further the theory of the optical properties of alloys so that estimates of the contribution of the k-nonconserving processes to $\Delta\sigma$ could be made.

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