Photoemission study of valence electrons in $La(Co_{1-x}Fe_x)_{13}$ (x=0,0.2)

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The electronic structure of $La(Co_{1-x}Fe_x)_{13}$ (x=0,0.2) has been investigated by using photoemission spectroscopy. The La 5d and T 3d (T=Fe,Co) partial spectral weights (PSW's) are extracted and compared with the calculated angular-momentum-projected densities of states (PLDOS's). No Co 3d satellite is observed, implying a weak Co 3d Coulomb correlation in this system. La 5d emissions exhibit a weak enhancement near the La 4d absorption edge, from which the La 5d PSW's of $La(Co_{1-x}Fe_x)_{13}$ (x=0,0.2) have been extracted. For a given x in $La(Co_{1-x}Fe_x)_{13}$, the overall features of the La 5d PSW, such as bandwidths and long tails, are rather similar to those of the T 3d PSW, indicating large hybridization between the T 3d and La 5d states. The experimental Co 3d and La 5d PSW's of $LaCo_{13}$ show a large disagreement with the calculated PLDOS's. With a substitution of Fe for Co, the PSW's of $La(Co_{1-x}Fe_x)_{13}$ remain nearly unchanged between x=0 and 0.2, which is contradictory to the predicted changes in the PLDOS's obtained in the rigid-band scheme. This finding suggests a breakdown of a simple rigid-band approximation in $La(Co_{1-x}Fe_x)_{13}$, which is probably due to a large hybridization bonding between Co and Fe 3d electrons.

I. INTRODUCTION

$LaCo_{13}$ has attracted much attention due to its potential application as a permanent magnetic material.[1–3] $LaCo_{13}$ has the largest 3d metal content among the known rare-earth (R)–transition-metal (T) intermetallic compounds, resulting in very large magnetization (1.3 T at room temperature) and a high Curie temperature T_C (T_C = 1318 K). Unfortunately, the possibility of using $LaCo_{13}$ as a permanent magnetic material has been low since its crystal structure is highly symmetric and so its magnetocrystalline anisotropy is small. Among the R–T binary systems (T = Fe, Co, Ni), only $LaCo_{13}$ crystallizes in a cubic $NaZn_{13}$-type structure.[1] That is, the $NaZn_{13}$-type structure is not stable for other R elements except for La, and neither $LaFe_{13}$ nor $LaNi_{13}$ exists.[4]

With an aim to find new permanent magnetic materials, many polycomponent systems based on $LaCo_{13}$ have been studied by replacing some of the Co in $LaCo_{13}$ with Fe, Al, and Ga, to produce materials such as $La(Co_{1-x}X_x)_{13}$ (X = Fe, Al, Ga) and $La(Co_{1-x-y}Fe_xAl_y)_{13}$.[5–7] Cubic $La(Co_{1-x}Fe_x)_{13}$ intermetallic compounds can be stabilized for $x \leq 0.6$.[7] In this system, T_C decreases and the magnetic moment per 3d atom increases, as x increases up to 0.6. On the other hand, the unique role of La in $LaCo_{13}$ in determining its structural stability and magnetic properties is not well understood yet, compared to other R elements. The effect of replacing La with other R elements in $LaCo_{13}$ has been studied, such as $La_{1-y}Nd_y(Co_{1-x}Fe_x)_{13}$ and $RCo_{13-x}Ga_x$ (R = La, Ce, Pr, Nd; $x \leq 4.4$).[7,8] At a certain range of x, the structural transformation was observed with a reduced T_C, but the lowering of symmetry in the cubic-tetragonal transformation did not result in a pronounced magnetocrystalline anisotropy.

The structural stability of a given material should be closely related to its electronic structure. In this aspect, it is important to understand the roles of the La 5d and Co 3d electrons in $LaCo_{13}$ in determining its structural stability and magnetic properties, as compared to other R and T elements.[9] In this paper we have investigated the electronic structure of $La(Co_{1-x}Fe_x)_{13}$ (x = 0, 0.2) by using synchrotron-radiation photoemission spectroscopy (PES) and compared PES results with band-structure calculations. This paper is organized as follows. In Sec. II, experimental and computational details are described. Section III is divided into three subsections. In Sec. III A, the photon energy $h\nu$ dependence of valence-band spectra of $La(Co_{1-x}Fe_x)_{13}$ are compared for x = 0 and 0.2. In Sec. III B, partial spectral weight (PSW) distributions of the La 5d and Co 3d electrons are extracted for x = 0 and 0.2, and compared with each other. In Sec. III C, experimental PES results are compared with theoretical density of states (DOS) and the correlation between the electronic structure and the structural stability in $LaCo_{13}$ is discussed. Finally conclusions are summarized in Sec. IV.

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II. EXPERIMENTAL AND COMPUTATIONAL DETAILS

$La(Co_{1-x}Fe_x)_{13}$ samples ($x = 0, 0.2$) were prepared by induction melting of the appropriate amounts of the constituent materials under pure argon atmosphere and then vacuum annealed at 1000 °C for one week. X-ray-diffraction measurements indicated that both samples were of single phase cubic $NaZn_{13}$-type structure with a negligible amount of impurity phases. The concentration of each element in $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$), which was checked by x-ray photoelectron spectroscopy, was very close to the nominal value, i.e., La: Co = 1: 13 for $x = 0$, and La: (Co + Fe) = 1: 13 and Co: Fe = 0.8: 0.2 for $x = 0.2$.

Photoemission experiments were performed on the Mark V beamline at the Synchrotron Radiation Center of the University of Wisconsin–Madison. Samples were cleaved *in situ* and measured at $\sim$ 60 K in a vacuum better than 4×10^{-11} Torr. A commercial double-pass cylindrical mirror analyzer (CMA) was used to analyze the kinetic energy (KE) of the emitted photoelectron. No carbon (C) or oxygen (O) contamination was detected in valence-band spectra as well as C and O 1*s* core level spectra. The overall instrumental resolution [full width at half maximum (FWHM)], due to a monochromator and the CMA, is 0.2 eV at $h\nu \simeq 40$ eV and 0.4 eV at $h\nu \geq 150$ eV, respectively. All the energy distribution curves (EDC's) were normalized to the incident photon flux. The Fermi level E_F of the spectrometer was determined from the valence-band spectrum of a sputtered Au sample.

Details of theoretical calculations have been described in an earlier paper.[9] The angular-momentum-projected local density of states (PLDOS) is obtained by using the linearized muffin-tin-orbital band method and the Gaussian broadening method. For the electron-electron exchange-correlation interaction, the von Barth–Hedin form is used in the local-density-functional approximation. Semirelativistic and self-consistent calculations were performed by including all the relativistic effects except for the spin-orbit effect. A ferromagnetic phase was assumed for $LaCo_{13}$ in order to compare the calculated PLDOS's with the experimental PES data which were taken at room temperature ($\ll T_C$). PLDOS's of $La(Co_{0.8}Fe_{0.2})_{13}$ were obtained by rigidly shifting E_F of PLDOS's of $LaCo_{13}$, taking account of a smaller number of valence electrons in $La(Co_{0.8}Fe_{0.2})_{13}$ than in $LaCo_{13}$.

III. RESULTS AND DISCUSSION

A. $h\nu$ dependence of the valence-band spectrum

Figures 1(a) and (b) show valence-band EDC's of $LaCo_{13}$ and $La(Co_{0.8}Fe_{0.2})_{13}$, respectively, around the Fe and Co 3*p* absorption thresholds. These EDC's are dominated by Co and Fe 3*d* emissions due to their large photoionization cross sections with respect to those of other electrons[10] and also due to a large number of T atoms (T = Fe, Co) per formula unit (f.u.) cell. Differences in EDC's of $LaCo_{13}$ and $La(Co_{0.8}Fe_{0.2})_{13}$ will reflect the effect of the Fe substitution for Co. As will be explained below, the $h\nu$ dependence of EDC's of $LaCo_{13}$ and $La(Co_{0.8}Fe_{0.2})_{13}$ greatly resembles each other. In both compounds, a broad Co *MVV* Auger emission is observed at a fixed KE, which appears at a binding energy (BE) of $\sim$ 7 eV for $h\nu = 60$ eV and shifts down through the valence band as $h\nu$ increases. Similarly, another weaker feature in $La(Co_{0.8}Fe_{0.2})_{13}$ at a lower KE (at BE $\sim$ 5 eV for $h\nu = 54$ eV) is due to the Fe *MVV* Auger emission. The intensity of the Co *MVV* emission is enhanced at $h\nu \simeq 60$ eV. The same phenomenon has been observed in Co metal[12,13] and in other metallic Co compounds.[11] The overall features of EDC's of $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$) are also rather similar to those of metallic Co compounds with smaller relative Co concentration.[11] Unlike other metallic Co systems,[11–13] however, no feature is observed in these compounds which might be identified as a "Co 3*d* satellite," indicating that the on-site Co 3*d* Coulomb interaction in $La(Co_{1-x}Fe_x)_{13}$ is weaker than that in pure Co metal[12,13] and in other metallic Co compounds.[11,14] Indeed, this indication is consistent with a smaller average interatomic distance between Co atoms and also with a smaller average magnetic moment per Co atom in $La(Co_{1-x}Fe_x)_{13}$ than in pure Co metal.[9]

Figures 2(a) and (b) show valence-band EDC's of

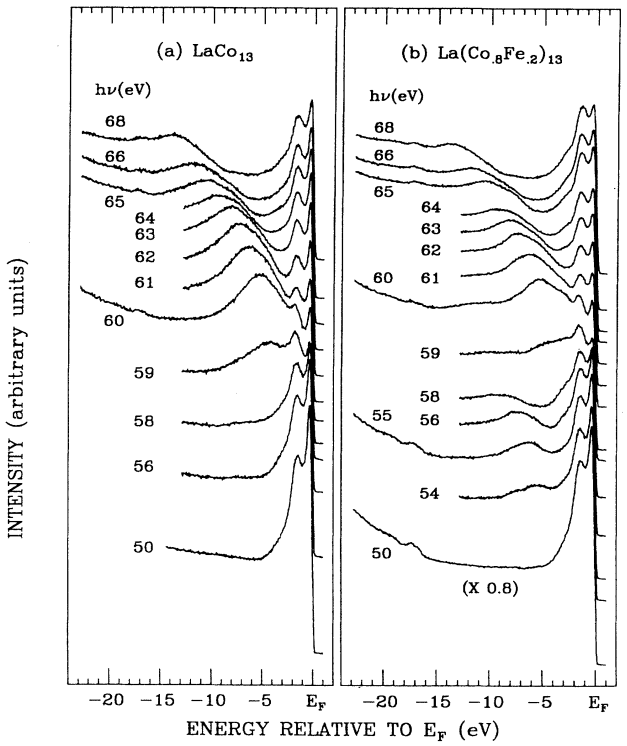

FIG. 1. (a) Energy distribution curves (EDC's) of $LaCo_{13}$ for $h\nu$ near the Co $3p \rightarrow 3d$ absorption edge, and (b) EDC's of $La(Co_{0.8}Fe_{0.2})_{13}$ for $h\nu$ including both the Co and Fe $3p \rightarrow 3d$ absorption edges.

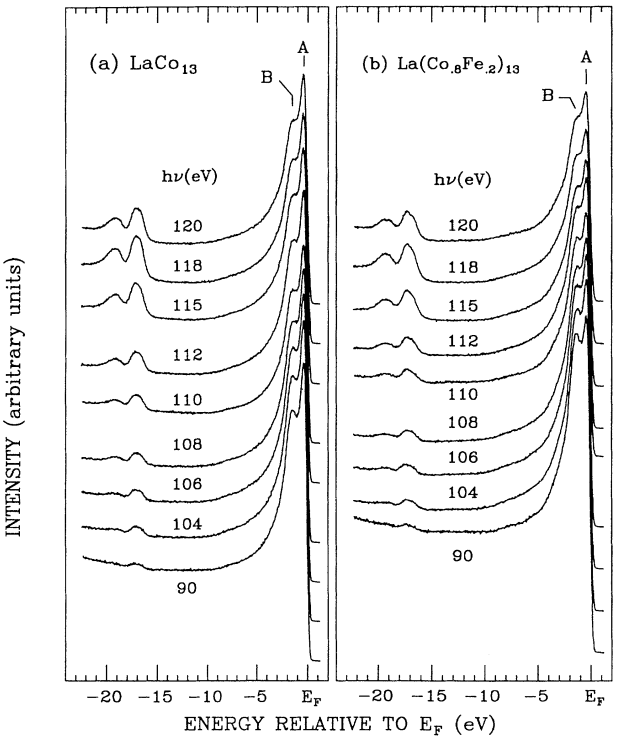

FIG. 2. (a) EDC's of $LaCo_{13}$ for $h\nu$ near the La $4d \rightarrow 4f$ absorption edge, and (b) similarly for $La(Co_{0.8}Fe_{0.2})_{13}$.

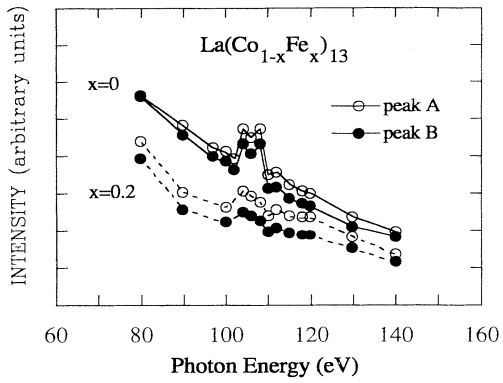

FIG. 3. Constant-initial-state (CIS) yield spectra of $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$), for two peaks A and B which are labeled in Fig. 2.

$LaCo_{13}$ and $La(Co_{0.8}Fe_{0.2})_{13}$, respectively, around the La $4d$ absorption threshold. The $h\nu$ dependence of EDC's is again very similar in both compounds: (i) as $h\nu$ varies, no pronounced changes are observed in the valence band region (between -5 eV and E_F), (ii) the La $5p$ emission at BE $\sim$ 18 − 20 eV exhibits a broad maximum near $h\nu \sim 110 - 120$ eV.[15,16] If La $5d$ emissions increase at certain $h\nu$'s, it will allow one to extract the La $5d$ PSW. By looking at these data, however, it is difficult to tell whether La $5d$ emissions are enhanced near the La $4d$ absorption edge, suggesting that the La $5d$ resonance is weak even if it exists. This finding is consistent with the delocalized nature of La $5d$ valence electrons in contrast to the localized R $5p$ and $4f$ electrons ($R \neq$ La).[16]

In Fig. 3, we show $h\nu$ dependences of the two peaks A and B, as labeled in Figs. 2(a) and (b). The $h\nu$ dependence of each spectral feature was obtained by estimating its amplitude above the inelastic background which was subtracted by using a conventional scheme.[17] The spectra in Fig. 3, obtained in this way, correspond to constant-initial-state (CIS) yield spectra. In both compounds, the intensities of the peaks "A" and "B" reveal an enhancement at $h\nu \sim 104$ eV, which might be identified as the Fano resonance in the La $5d$ photoionization cross section, similar to that seen in some other rare-earth metals.[16] Then the resonance energy of the La $5d$ cross section ($h\nu \sim 104$ eV) is well below that of the La $5p$ cross section ($h\nu \sim 110 - 120$ eV). Note that the Fano resonance involves the quantum interference between the direct photoemission process and the Auger autoionization process subsequent to the core-level photon absorption.[15] The resonance energy is determined by the magnitude of the Auger matrix element between the discrete intermediate state and the continuum final state, which causes a shift with respect to the absorption edge. Due to the localized nature of the La $5p$ electrons, as compared to the La $5d$ electrons, it is expected that the Auger matrix element for La $5p$ emissions is larger than that for La $5d$ emissions.[18] This will give rise to a larger energy shift in the Fano resonance energy of the La $5p$ cross section, than that of the La $5d$ cross section.

B. La 5*d* and Co 3*d* partial spectral weight (PSW)

Figure 4(a) presents the La $5d$ PSW's in $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$), which were extracted by subtracting the spectra at $h\nu = 100$ eV from those at $h\nu = 104$ eV corresponding to the Fano resonance. Here we have assumed that La $5d$ emissions in the $h\nu = 100$ eV spectra are negligible, since Fe and Co $3d$ emissions are dominant off resonance.[10,19] This figure shows that the La $5d$ PSW's can be separated from the T $3d$ PSW's in $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$). To our knowledge, the extraction of the La $5d$ PSW's using resonant photoemission has not been reported before for La compounds. The extracted La $5d$ PSW's are rather noisy, compared to the T $3d$ PSW's in Fig. 4(b). This is because the resonance in the La $5d$ cross section is weak and the number of La atoms per f.u. is small with respect to that of T atoms.

In Fig. 4(b), we have chosen the spectra at $h\nu = 90$ eV as representing the T $3d$ PSW's of $La(Co_{1-x}Fe_x)_{13}$. As pointed out above, non-T $3d$ emissions are negligible in the region of 70 eV $< h\nu <$ 100 eV.[19,20] This is indeed reflected in Figs. 1 and 2 by the fact that the EDC line shapes hardly change as $h\nu$ varies from 70 to 100 eV. In Fig. 4(b), spectra for two compositions are scaled so that the area underneath each spectrum is proportional to the average number of the $3d$ electrons per T atom, n_{3d}. For $LaCo_{13}$, $n_{3d} = 7.54$ was used, which was obtained from band-structure calculations. For $La(Co_{0.8}Fe_{0.2})_{13}$,

n_{3d} was determined from a rigid shift in E_F of $LaCo_{13}$. Substitution of 20% of Fe for Co in $LaCo_{13}$ caused a shift in E_F of 0.15 eV,[21] resulting in $n_{3d} = 7.36$.

Figure 4 reveals that there is no overall shift in the La $5d$ and T $3d$ PSW's of $La(Co_{1-x}Fe_x)_{13}$ between $x = 0$ and 0.2. Overall features of the La $5d$ PSW barely change with a substitution of Fe for Co in $LaCo_{13}$, which is consistent with the delocalized nature of the La $5d$ electrons. Similarly, the FWHM of the T $3d$ PSW remains unchanged as x increases from 0 to 0.2. Further, there is no shift in peak positions within the experimental resolution. The only change observed is a transfer of the spectral weight from the near E_F region to the shoulder at ~ -1.5 eV. These findings suggest that the rigid-band approximation might not work for describing electronic structures of $La(Co_{1-x}Fe_x)_{13}$.

This figure provides other important information on $La(Co_{1-x}Fe_x)_{13}$. For a given x, the La $5d$ and T $3d$ PSW's of $La(Co_{1-x}Fe_x)_{13}$ share some features in common. First, most of the spectral weights are concentrated in the region between ~ -3 eV and E_F. Second, the full widths extend from E_F to $5 \sim 6$ eV below E_F, and so the La $5d$ and T $3d$ PSW's occupy nearly the same energies. Finally, the line shape of the La $5d$ PSW's of $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$) is very different from that of La metal.[20] These findings indicate that the hybridization interaction between the La $5d$ and T $3d$ states is large in $La(Co_{1-x}Fe_x)_{13}$.

C. Comparison of PES spectra with band-theoretical results

We have calculated the La $5d$ and T $3d$ PLDOS's of $La(Co_{1-x}Fe_x)_{13}$ (x=0,0.2), motivated by the lack of a Co $3d$ satellite structure in $LaCo_{13}$ (see Fig. 1). For Co metal and some metallic Co systems, a reasonably good agreement has been observed between experimental valence-band PES spectra and band-theoretical calculations, except for some discrepancies which are considered to arise from the effects of the on-site Co $3d$ Coulomb correlation[11,22] and photoionization matrix elements.[11] Thus it will be worthwhile to compare experimental data with band-theoretical results for $La(Co_{1-x}Fe_x)_{13}$.

Figures 5(a) and (b) compare the calculated PLDOS's of the La $5d$ and T $3d$ states of $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$), respectively. The PLDOS's of $La(Co_{0.8}Fe_{0.2})_{13}$ were obtained by a rigid-band shift of E_F downward from those of $LaCo_{13}$ by $\sim$ 0.15 eV, to account for a smaller number of valence electrons in $La(Co_{0.8}Fe_{0.2})_{13}$. As a consequence of a rigid shift in E_F, the following changes are observed in the PLDOS's with $x = 0.2$, compared to those with $x = 0$: (i) peak positions move toward E_F, (ii) occupied bandwidths decrease. These theoretical trends do not agree with those of the experimental PSW's in Fig. 4, indicating that a simple rigid-band approximation in obtaining the PLDOS might not work for $La(Co_{1-x}Fe_x)_{13}$. The

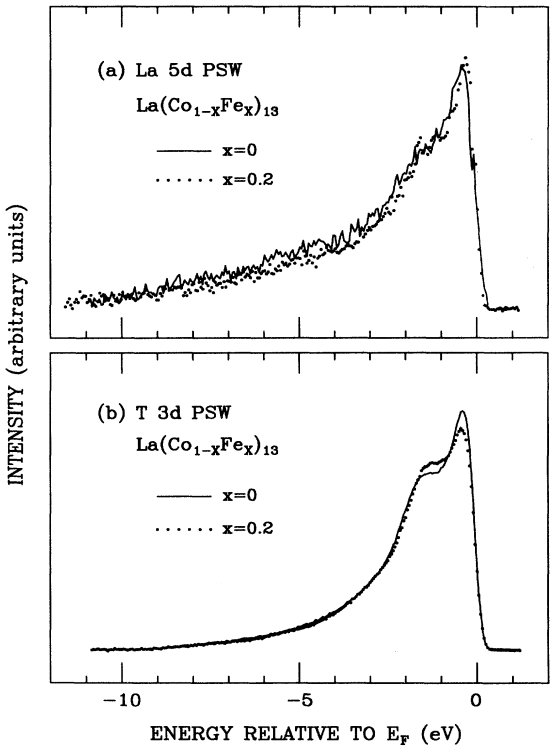

FIG. 4. (a) Comparison of the extracted La $5d$ partial spectral weight (PSW) of $LaCo_{13}$ with that of $La(Co_{0.8}Fe_{0.2})_{13}$. (b) Similarly for the T $3d$ PSW's (T=Co,Fe).

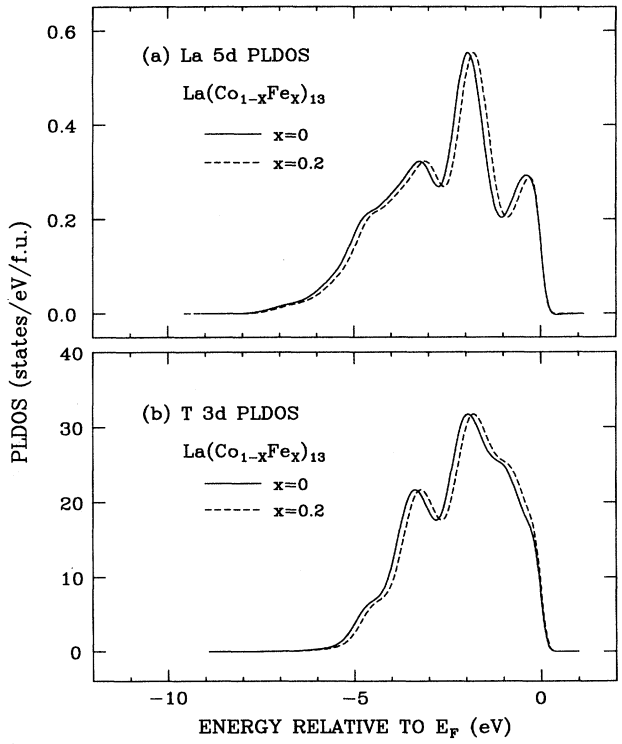

FIG. 5. (a) Comparison of the calculated angular momentum projected local density of states (PLDOS) for the La $5d$ states in $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$). (b) Similarly for the T $3d$ PLDOS (T=Co,Fe).

reason for the breakdown of the rigid-band model for $La(Co_{1-x}Fe_x)_{13}$ seems to be due to a large hybridization bonding between Co and Fe 3*d* orbitals, which will result in a substantial modification of the DOS structure.

There are two more points to consider. First, for a given x in $La(Co_{1-x}Fe_x)_{13}$, several structures in the La 5*d* PLDOS are common to those in the *T* 3*d* PLDOS, suggesting that there is large hybridization between the La 5*d* and *T* 3*d* electrons in $La(Co_{1-x}Fe_x)_{13}$. This is in accord with the experimental finding in Fig. 4. Second, the scale of the La 5*d* PLDOS is smaller than that of the *T* 3*d* PLDOS by about two orders of magnitudes. Note that other *R* elements ($R \neq$ La, *R*:Ce–Lu) have large *R* 4*f* PLDOS's,[11,16] which are expected to overlap energetically with the *T* 3*d* PLDOS.[11,14,23,24] Concomitant substantial *f*-*d* hybridization may result in a high DOS at E_F, which then gives rise to a structural instability. Thus it is speculated that the unstable structure of RCo_{13} for $R \neq$ La might be related to the presence of the large 4*f* PLDOS in the region of the Co 3*d* PLDOS.

Figures 6(a) and (b) compare the experimental La 5*d* and Co 3*d* PSW's with the calculated La 5*d* and Co 3*d* PLDOS's of $LaCo_{13}$, respectively. It is clear that calculated PLDOS's and experimental PSW's do not agree in details. Part of these discrepancies might be due to the effects neglected in the theory curves, such as the effect of photoionization matrix elements[25,26] and the relaxation effect in photoemission processes. If the effect of finite lifetimes of valence holes[27] is included in the calculated Co 3*d* PLDOS, which is also neglected at present, even a larger discrepancy is expected between theoretical and experimental FWHM's for Co 3*d* states. It is surprising that the discrepancies between theory and experiment in $LaCo_{13}$ are larger than those in Co metal and some other Co-based metallic systems.[11,22] Usually such a breakdown in agreement between one electron band-theoretical results and photoemission data have been attributed to the effects of electron-electron correlations. For $LaCo_{13}$, however, that does not seem to be the case because of the lack of a Co 3*d* satellite. The orgin of this disagreement between theory and experiment needs to be further investigated.

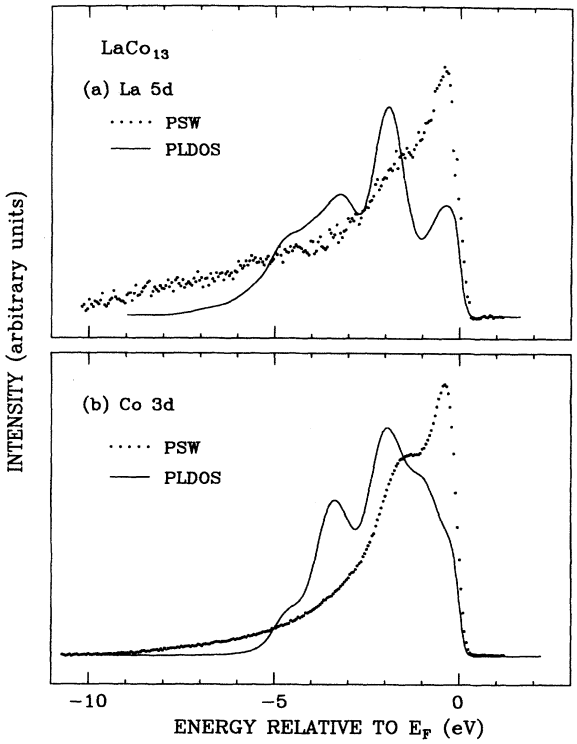

FIG. 6. (a) Comparison of the experimental La 5*d* PSW with the calculated La 5*d* PLDOS of $LaCo_{13}$. (b) Similarly for Co 3*d* states.

IV. CONCLUSIONS

Using synchrotron-radiation photoemission spectroscopy, we have investigated electronic structures of $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$), which have the highest 3*d* metal content among the known rare-earth intermetallic compounds. Using the $h\nu$ dependence of La 5*d* and *T* 3*d* emissions (*T*=Fe,Co), we have determined the La 5*d* and *T* 3*d* PSW's. It is found that the overall features of valence-band spectra of $La(Co_{1-x}Fe_x)_{13}$ with $x = 0.2$ are very similar to those with $x = 0$ and also to those of metallic Co compounds with a smaller relative Co concentration. A weak enhancement has been observed in the La 5*d* photoionization cross section near the La 4*d* absorption edge, from which the La 5*d* PSW's of $La(Co_{1-x}Fe_x)_{13}$ ($x = 0, 0.2$) are extracted. With substitution of Fe for Co in $LaCo_{13}$, there is no overall shift in the La 5*d* and *T* 3*d* PSW's, except for small spectral weight transfer in the *T* 3*d* PSW's. For a given x in $La(Co_{1-x}Fe_x)_{13}$, the overall features of the La 5*d* PSW greatly resemble those of the *T* 3*d* PSW, suggesting large hybridization between the *T* 3*d* and La 5*d* states. This is also supported by the calculated La 5*d* and Co 3*d* PLDOS's.

Unlike in pure Co metal and other metallic Co compounds, no Co 3*d* satellite feature is observed in $LaCo_{13}$, implying a weaker on-site Co 3*d* Coulomb correlation in $LaCo_{13}$. However, the calculated Co 3*d* and La 5*d* PLDOS's for $LaCo_{13}$ exhibit large discrepancies from experimental PSW's, in contrast to the expectation based on the lack of a Co 3*d* satellite. Further, the predicted changes in PLDOS's with a substitution of Fe for Co in $LaCo_{13}$, obtained in the rigid-band approximation, are found to be contradictory to those of experimental PSW's. This finding suggests that a simple rigid-band approximation might not work to describe the electronic structure of $La(Co_{1-x}Fe_x)_{13}$ for $x < 1$, probably due to a large hybridization bonding between Co and Fe 3*d* orbitals. It is also speculated that the instability of a RCo_{13}-type structure for other *R* elements ($R \neq$ La) might be related to high *R* 4*f* DOS's which overlap energetically with Co 3*d* states.

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