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Electronic structure of α- and δ-Pu from photoelectron spectroscopy

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We report photoemission results on α- and δ-Pu using a laser plasma light source (LPLS) as well as He light as the exciting radiation. The LPLS is a pseudocontinuum tunable light source with intensities rivaling some second-generation synchrotrons. Both phases of Pu display a narrow, temperature-independent, $5f$-related feature at the Fermi energy, which is narrower in δ-Pu than in α-Pu, suggestive of possible heavy-fermion-like behavior. In both α-Pu and δ-Pu the photon-energy dependence of this feature suggests some $6d$ admixture, albeit somewhat smaller in δ-Pu. In this respect it differs qualitatively from Ce and U heavy fermions.

I. INTRODUCTION

The electronic structure of the $5f$ series of metals in the periodic table represents one of the last unexplored areas in condensed matter physics. Indeed, it probably represents the most interesting area of materials properties since here the $5f$ electrons tend to behave in a highly correlated manner as opposed to the single-particle behavior so prevalent in simpler systems. The myriad of narrow-band phenomena,[1,2] ranging from magnetism through heavy-fermion behavior to normal transition metal behavior, is unparalleled in any other series of elements.

The metal plutonium appears to be the most exotic of all, displaying six allotropic phases[3] and forming the boundary between localized (Am) and delocalized (Np) behavior of the $5f$ electrons.[4] The resistivities of both α- (Ref. 5) and δ-Pu (Ref. 6) are large and have a negative temperature coefficient,[7] which is highly unusual for an elemental material. It is the delicate balance between $5f$ bonding and localization that is most likely responsible for many of plutonium's unusual properties, including its propensity for phase changes, low-symmetry crystal structures, and low melting point. By virtue of its structural diversity, Pu presents a unique opportunity to observe the modifications in electronic structure effected by the allotropes' striking volume and structural differences.

The low-temperature phase of Pu (α-Pu) is characterized by a monoclinic crystal structure[8] and a high density ($\sim$19 g/cm^3). Local-density-approximation (LDA) band calculations[9] suggest that one should obtain direct f-f wave-function overlap, yielding narrow but robust $5f$ bands so that the material can be described as having transition metal properties. The high-temperature δ phase, by contrast,[10] is a fcc structure with a density of $\sim$16 g/cm^3. Direct f-f overlap is considered unlikely for most of the $5f$ electrons. Small residual hybridization, as per the periodic Anderson model,[11] or perhaps itinerancy from one out of the five Pu $5f$ electrons,[12] may yield some $5f$ density of states (DOS) at E_F. Indeed, recent magnetic susceptibility measurements suggest that δ-Pu is a heavy-fermion material.[13]

Heavy fermions are metals or compounds with an unfilled $4f$ or $5f$ shell whose bulk properties suggest[14–16] that the f electrons at the Fermi energy display a crystal effective mass that is orders of magnitude larger than the rest mass of the electron and, indeed, much larger than the calculated band mass for the material.[15] A long-standing (though somewhat simplistic) model to explain this behavior is the single-impurity model (SIM),[17,18] which postulates that the f electrons behave as noninteracting magnetic impurities on every atomic site at high temperatures. As the temperature is lowered, a cloud of non-f conduction electrons is postulated to align antiparallel to the f spin to form a singlet state, thus quenching the magnetic moment below a characteristic temperature called the Kondo temperature, or T_K. Within the SIM, very slight residual f-electron hybridization with this compensating cloud produces a narrow resonance near the Fermi energy (at k_BT_K above E_F for a less than half filled f shell, and k_BT_K below E_F for more than half filling), referred to as the Kondo resonance, which is expected to yield a very narrow sharp feature at E_F in a photoelectron spectroscopy (PES) spectrum. However, the SIM applicability to periodic systems is tenuous at best since the concept of an impurity on every site is almost a contradiction in terms.[19,11] Recent measurements in uranium heavy-fermion compounds[20,21] suggest that LDA-derived bands are well reproduced experimentally, except at the Fermi energy where they are flatter and narrower than the calculated bands. A comprehensive model that incorporates both lattice periodicity as well as electron correlations is yet to be worked out.[11] Unwarranted adherence to any one particular model could impede progress in the $5f$ series. Clearly, a detailed understanding of the Pu $5f$ electronic structure is of fundamental importance to the overall understanding of properties of Pu, and to condensed matter physics as a whole.

Photoelectron spectroscopy represents a direct and detailed tool for experimentally determining the electronic structure of a material. This is especially true when this tool is used in conjunction with a tunable light source where it is possible to select any desired photon energy, owing to the fact that the cross section[22] for photon absorption is a strong function of both electron orbital symmetry and photon en-

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ergy. In this work we make use of a benchtop tunable light source,[23] the laser plasma light source (LPLS), to deconvolve the orbital character via tunable light. The LPLS is the only benchtop tunable light source for valence-band PES dedicated to transuranic research. Spectra using photon energies of 21.2 and 40.8 eV represent He I and He II radiation, respectively, and are an integral part of the LPLS system. These energies are used for the higher-resolution spectra below.

II. EXPERIMENTAL DETAILS

The hazardous nature of Pu generally precludes the use of synchrotrons for photoelectron spectroscopy except for some limited applications. Although some Pu experiments are now possible owing to the undulator intensities and hence the rapidity with which spectra can now be obtained, most experiments still require *in situ* cleaning of the surface, particularly for work at low photon energies where, in the presence of surface oxygen, the O cross section dominates.[22] In order to circumvent *in situ* cleaning difficulties (at public facilities) associated with highly radioactive materials, we have developed a tunable light source,[23] the LPLS, whereby we are able to mimic the output of a synchrotron within our own laboratory with a benchtop system, albeit at lower intensities. In the LPLS apparatus a KrF excimer laser pulse of 248 nm light and 20 nsec duration is focused onto a target consisting of a thin stream of Hg, thus creating a plasma. The soft x rays emitted by plasma recombination are collected (30×100 mrad) and monochromated using a Hettrick in-focus variable-groove-spacing monochromator, with a maximum resolving power of 1000. Light intensity is measured as a photocurrent from a 90% transmitting Ni mesh, with typical currents of 1–5 pA at maximum resolving power.

A VSW HAC100 electron energy analyzer is used for energy analysis of the photoemitted electrons. The electron detector was modified with a multichannel plate and phosphorescent screen, while a charge-coupled device (CCD) camera recorded the electron events on the phosphorescent screen. The use of a CCD camera is essential owing to the time structure of the incoming light (typically 50 Hz at 20 nsec per pulse, although the system is capable of 200 Hz). It circumvents the dead time associated with resistive anode detectors. With the use of the He lamp the best resolution is typically 50 meV, while with the LPLS, resolutions of 150 meV are more common. Measurements were performed at either 300, 80, or 20 K.

The Pu specimens were polycrystalline samples typically $3\times3\times1$ mm^3, and weighing about 200 mg. These were soldered onto Cu posts and inserted into the spectrometer via a load-lock transfer system. We measured both aged (about 10 yr) and freshly prepared specimens, with the Ga-stabilized δ-Pu specimens having a nominal Ga concentration of 1 wt %.

Surface preparation is a crucial part of a PES experiment since the probing depth of PES is only a few atomic layers,[24] thus requiring optimal coupling between the surface and bulk wave functions to accurately measure the bulk. Atomically clean surfaces are essential. With the in-house LPLS system we can redirect the laser beam into the vacuum chamber through a quartz window to clean the sample surface via laser ablation. The advantage of this is that it is trivial to perform cleaning in the vacuum through a quartz window starting with a ‘‘dirty’’ sample, and it can be done in seconds while the sample is at cryogenic temperatures. The low temperature reduces the migration of impurities from the bulk onto a freshly cleaned surface. The ablation technique is demonstrated in Fig. 1 where PES spectra from an α-Pu specimen are shown in various stages of cleaning in the spectrometer chamber, starting with primarily a PuO$_2$ surface. PES data taken at a photon energy of 40 eV (note, not 40.8 or He II) are both very surface sensitive and at a maximum for O $2p$ photoelectron cross section.[22] This enhances the oxygen signal from the surface oxygen relative to the Pu in the bulk. In Fig. 1 only a few laser pulses represent each cleaning. Note that with each cleaning the Fermi edge grows in intensity, while the oxide peak at -6 eV binding energy decreases and changes in line shape (residual surface oxygen is still present in cycle 5, but the line shape is clearly different from the starting oxide, which suggests adsorbed oxygen). Using these techniques we have obtained PES data on α- and δ-Pu at a number of different photon energies. The higher resolutions (relative to previous work[25,26]) and cleaner surfaces have yielded dramatic, unexpected results as we show below.

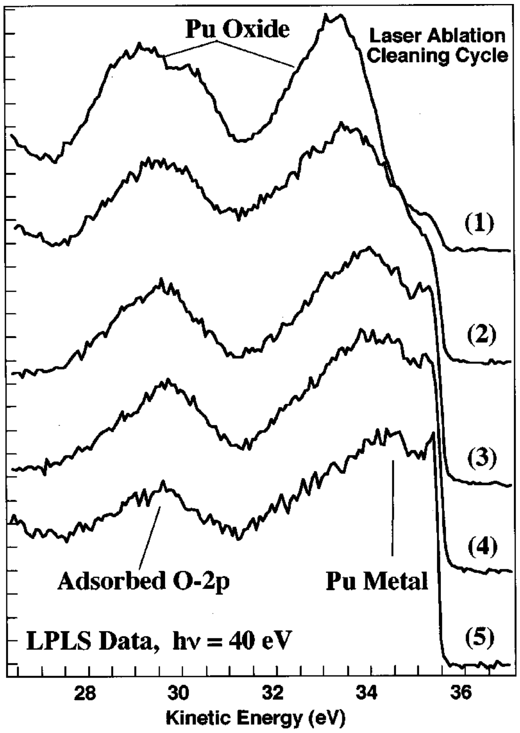

FIG. 1. Spectra from various stages of cleaning of α-Pu via laser ablation, starting with a Pu oxide surface. Note the change in line shape of the oxygen feature at 6 eV below E_F, and the growth of the Fermi edge with progressive ablation. In cycle 5 the O $2p$ signal is probably due to adsorbed oxygen.

III. PES RESULTS AND DISCUSSION

In Fig. 2(a) we show energy distribution curves (EDCs) for the two Pu allotropes using He II radiation with the specimens at 80 K. By collecting the data at 80 K and working at a photon energy of 40.8 eV where the $6d$ and $5f$ cross sec-

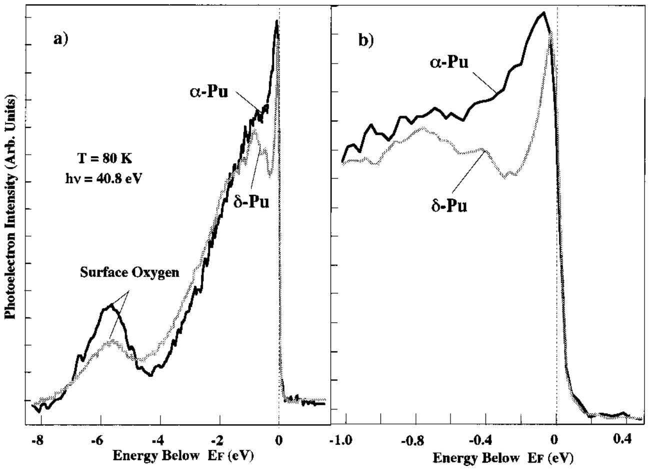

FIG. 2. (a) Wide-scan spectra of α- and δ-Pu at 80 K using He II radiation. The feature at -6 eV is due to about 0.1 monolayer of adsorbed oxygen. The spectra nearly overlie each other below ~ -1 eV. (b) Expanded view of spectra near the Fermi energy. The significant difference between them is the broader nature of the near-E_F feature in α-Pu.

tions per electron are nearly equal,[22] we obtain, perhaps, a good representation of the DOS in the PES spectra. In Fig. 2 a Shirley-type background has been subtracted and the spectra normalized to equal spectral weights between E_F and -4 eV. With the exception of the feature near the Fermi energy, the spectra are surprisingly similar (given the volume change and differences in mechanical properties and crystal structure). There are also some very subtle and perhaps insignificant intensity differences in the -1.5 eV range. The intensity at about -6 eV is due to adsorbed oxygen from the edge portions of the sample not fully cleaned by ablation. The sharp, near-E_F structure for δ-Pu is more intense and narrow than that of α-Pu, consistent with a narrower DOS and hence greater tendency toward localization.

Concentrating on the expanded view in Fig. 2(b) where only the near-E_F data are shown, we find the only truly significant and understandable differences between α-Pu and δ-Pu in the valence-band data. In particular, the sharp feature in δ-Pu [full width at half maximum (FWHM) $\leqslant 100$ meV] is clearly much narrower than that found in α-Pu (FWHM $\geqslant 200$ meV) and, to within our resolution, crosses E_F, while the much broader peak in α-Pu reaches maximum intensity at about 100 meV below E_F. This is consistent with the larger specific heat γ value of δ-Pu,[27] relative to that of α-Pu. Comparison of this spectrum to spectra in uranium compounds suggests that δ-Pu could be identified as a heavy fermion.[18] However, there are differences, as described below.

Data taken at 21.2 eV light (He I radiation) are shown in Fig. 3 for both α-Pu and δ-Pu, where across the full valence band both PES spectra again appear surprisingly similar except for the near-E_F feature which is, as in the 40.8 eV data, much narrower and somewhat more intense in δ-Pu. Since for the most part the data are similar to data presented in Fig. 2, despite the fact that the $6d$ cross section should predominate over the $5f$ at 21.2 eV, this suggests that there exists some $6d$ character at E_F and that $6d$ features display a narrowness similar to the $5f$ states.

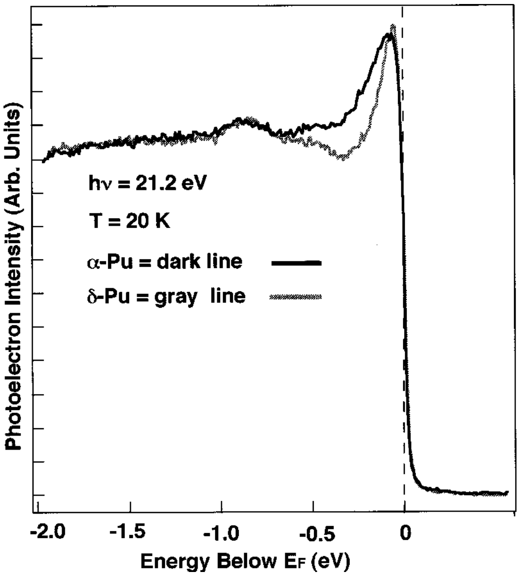

FIG. 3. Spectra for α- and δ-Pu at 21.2 eV and 80 K. Again the spectra overlie each other almost exactly except near the Fermi energy. Here the spectra should be dominated by $6d$ character, yet most features are similar to those at higher energies. The $6d$ weight may be spread throughout the valence-band region.

Because of the similarity of the α- and δ-Pu spectra there has been speculation[28,29] that the surface of α-Pu may contain a δ-like structure. However, in Fig. 4 we show α-Pu spectra taken with 21.2, 30, 40.8, and 60 eV light. Using conventional escape depth arguments,[24] the low-energy data should be more representative of the bulk, yet all features are present at all energies. The sharp feature at E_F is in fact

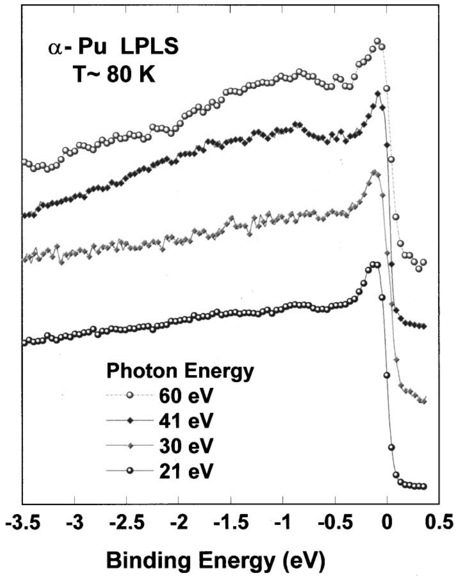

FIG. 4. Spectra of α-Pu at 80 K taken at the various indicated photon energies. The narrow feature near E_F is persistent at all photon energies and must thus contain substantial $6d$ admixture in its orbital symmetry, since at 21.2 eV it should be dominated by $6d$ emission (see Fig. 5). Moreover, it is most likely bulk derived based on the larger escape depth at 21.2 eV. The purer $5f$ states are in the -1 eV region which clearly grows faster with photon energy than the near-E_F peak.

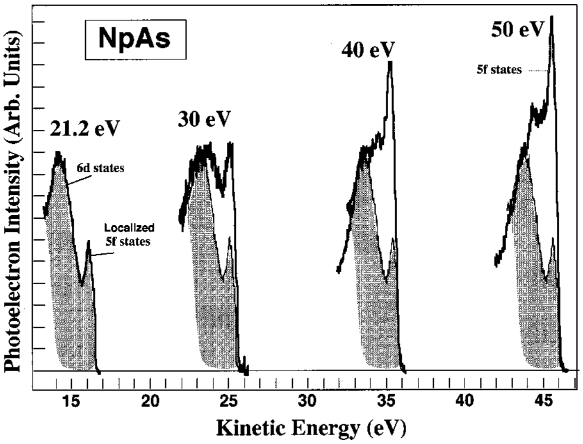

FIG. 5. Spectra of NpAs are displayed at the indicated photon energies. The spectrum taken with $h\nu = 21.2$ eV is of primarily $6d$ character. It has been superimposed on the remaining spectra to highlight the growth of the $5f$ features. Note the difference in line shape due to increased $5f$ emission at higher $h\nu$.

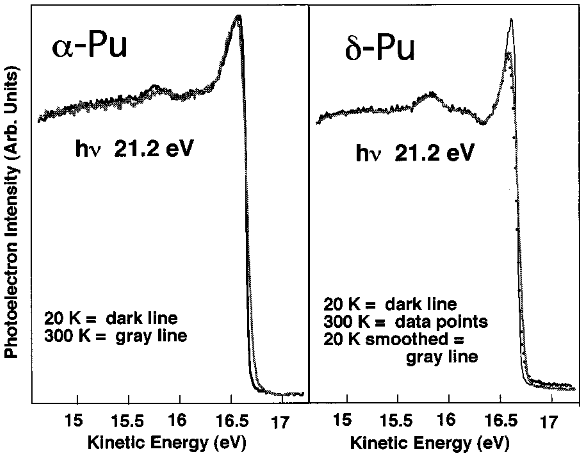

FIG. 6. Spectra of α-Pu (left panel) and δ-Pu (right panel) at 20 and 300 K. α-Pu is temperature independent, while the small temperature dependence of δ-Pu can be completely accounted for by a convolution with a 110 meV FWHM Gaussian which approximates the Fermi function broadening at 300 K.

somewhat more intense relative to the other features (centered at about -1.2 eV) at 21.2 eV photon energy than at higher energies, which indicates not only that this peak derives from the bulk, but also that it contains more $6d$ admixture in its orbital character than the remaining features, which may be of purely $5f$ character. From Figs. 2 and 3 one can deduce that in δ-Pu we are also dealing with bulk phenomena in that the 21.2 eV data are similar to 40.8 eV data, although now there appears to be a smaller change in the intensity of the sharp peak at E_F relative to other features. Very similar data[36] at a photon energy of 220 eV likewise support the bulk-derived interpretation of spectral features. This suggests that the orbital mix of the near-E_F peak in δ-Pu contains somewhat more $5f$ character than in α-Pu. This would be consistent with the conventional wisdom that α-Pu is more hybridized and bandlike than δ-Pu.

The escape depth argument is obscured somewhat by the possibility of $6d$ admixture, as well as the fact that the $5f$ cross section varies dramatically in this range.[22] For this reason we show in Fig. 5 similar data in NpAs where the $5f$ electrons are most likely localized and the system is in the antiferromagnetic state at 80 K, in order to demonstrate the cross-sectional differences between $5f$ and $6d$ electrons. Here the $5f$ and $6d$ features are well separated and one can clearly discern the growth of the $5f$ peaks with photon energy relative to the $6d$. The sharp $5f$ peak is situated about 0.4 eV below E_F (localized) with weak intensity at $h\nu = 21.2$ eV. The 21.2 eV spectrum has been superimposed on the higher-energy spectra for ease of viewing the $5f$ intensity increase relative to the $6d$ states. This photon-energy sequence confirms that we are observing bulk phenomena at these energies. At $h\nu = 40$ eV the $5f$ intensity has already surpassed the $6d$ intensity. Since the integrated $6d$ content in Pu is expected to be similar to that of Np, a reasonable conclusion again follows that the sharp near-E_F features in Pu must contain some $6d$ admixture, and the $6d$ weight is spread throughout the spectrum, otherwise the near-E_F peak would be much smaller at 21.2 eV relative to other features, as it is in NpAs.

We now contend with the fact that both allotropes of Pu apparently exhibit very narrow bands at E_F and very similar electronic structures at higher binding energies. While very-narrow-band behavior was in fact anticipated for δ-Pu (and is obtained in a band calculation; see below[12]) it is unexpected for α-Pu where all indications from bulk measurements[5,7] are that the $5f$'s are strongly hybridized with the $6d$'s and indeed may have direct *f*-*f* overlap. While the near-E_F feature in α-Pu is twice as broad as that in δ-Pu and reaches a maximum intensity 100 meV below E_F, it is nevertheless not reproduced in a band calculation (see below). This, combined with the overall similarities between the two spectra at higher binding energies, precludes a complete understanding at this point.

Invoking Kondo-like electron-electron correlations (e.g., the single impurity model[17,18]) even in α-Pu as the source of the narrow structure presents problems. The fact that the maximum in intensity occurs clearly below E_F, combined with the indication of strong *d*-*f* admixture, puts such an interpretation in doubt. A SIM-like DOS in a less than half filled *f* shell should be positioned above the Fermi energy with only its tail extending into the occupied states.[17,18] Thus in Pu allotropes the maximum PES intensity must, of necessity, be at E_F within such an interpretation. Similar problems are not encountered in δ-Pu where the PES spectrum actually exhibits a maximum intensity precisely at E_F (to within our resolution) and is thus partially consistent with the above interpretation. For this reason the SIM interpretation needs further testing.

One of the tests for the applicability of the SIM to Pu is a determination of the temperature dependence[18] of the narrow $5f$ features. In Fig. 6 we show data taken at 21.2 eV at both 300 and 20 K for both α-Pu and δ-Pu. No change with temperature is evident in α-Pu (consistent with no electron-electron correlations attributable to SIM), while a decrease in intensity actually occurs at 300 K in δ-Pu. One must, however, take into account the role of the Fermi function truncating this narrow feature prior to ascribing a temperature dependence to the effects of correlations within the SIM or any other model. A 110-meV-wide Fermi function (i.e., $4.4k_BT$ where $T \approx 300$ K) can be approximated by convolut-

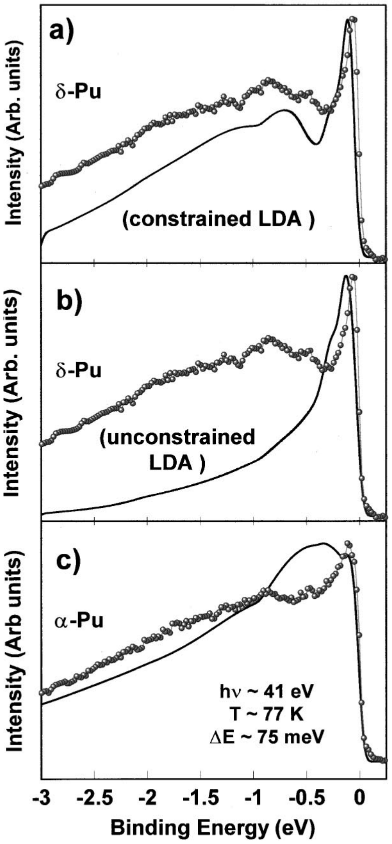

FIG. 7. Comparison of spectra (data points) to band calculations (solid curves). Calculated curves have been broadened as indicated in the text while a Shirley background has been subtracted from the data. (a) δ-Pu compared to a constrained LDA calculation where only one $5f$ electron is itinerant. (b) δ-Pu compared to an unconstrained LDA calculation. The comparison is very poor. (c) α-Pu compared to an unconstrained LDA calculation. Sharp feature at E_F is not reproduced.

ing the spectrum with a 110-meV-wide Gaussian, since the remaining broad features will be unaffected by this convolution. When this is done, all the temperature dependence between 20 and 300 K is accounted for, thus making it unnecessary to invoke additional phenomena. We realize that this is only an approximation, but it immediately yields the bulk of the answer.

Comparison to band calculations is shown in Fig. 7 for both α-Pu and δ-Pu. In each frame the calculated DOS (solid curves) has been convoluted with a Gaussian whose full width at half maximum increases as $[E_0+E_1(E_B^2)]$, where E_B is the energy below the Fermi energy, E_0 is set to 20 meV, and E_1 is 1.1 $(\mathrm{eV})^{-1}$ to simulate lifetime broadening of the photohole.[30] The maximum allowed broadening[31] is restricted to 1 eV. All calculations are broadened identically. The secondary electrons have been subtracted out of the experimental data via a Shirley-type background subtraction.[24]

In Fig. 7(a) the δ-Pu spectrum is compared to a constrained LDA calculation[12] where four of the five $5f$ electrons are artificially constrained to localization. Globally the experimental δ-Pu spectrum is well reproduced by the calculated spectrum, suggesting that perhaps this could be a starting point for a basic understanding of Pu. This notion is reinforced by the observation that the band calculations predict some admixture of $5f$ and $6d$ character for the narrow peak, the amount depending on the choice of constraint conditions. The photon-energy dependence of the data suggests that such a mix is reasonable for δ-Pu, as dicussed above. However, the calculation at the present time would place the narrow feature a few meV below E_F as opposed to exactly at E_F where it is found experimentally (a 60 meV shift would overlay the data). The exact nature of this discrepancy is not understood at this time.

In Fig. 7(b) we compare the δ-Pu spectrum to an unconstrained LDA calculation where all $5f$ electrons are allowed to be itinerant. Within this calculation the bulk of the $5f$ DOS is found to be contained in a narrow region within 0.5 eV of the Fermi energy (similar to that of uranium). This is clearly at odds with experiment, so that the the unconstrained calculation misses badly. It reinforces the notion of localization for at least some of the $5f$ electrons.

The α-Pu calculation in Fig. 7(c) would seem to present more serious problems since the sharp structure at E_F is not well reproduced in an unconstrained LDA calculation. The occurrence of this feature at 100 meV below E_F would seem to rule against correlation effects. However, while one would not anticipate electron-electron correlations in a system with a temperature-independent susceptibility[5] as well as a dense structure where f-f overlap is possible, one cannot neglect the anomalous resistivity[5] which suggests unusual scattering behavior. Moreover, as we state below, core-level spectra[32] show satellite behavior even in α-Pu. Perhaps correlation phenomena are important in spite of the $5f$ maximum occurring at 100 meV below E_F. A very reasonable calculational approach might be one of renormalized bands in which the Hubbard Hamiltonian is introduced as a perturbation on LDA-derived bands. By varying the Coulomb correlation U one controls the strength of the electron-electron correlations. In this way both α- and δ-Pu may yet be understood.

The intense radioactivity of Pu introduces radiation damage and daughter products over time in the Pu matrix. The radiation damage tends to anneal out even at room temperature[33] and is in any case eliminated by the laser ablation. However, the buildup of impurities may have an effect on the α-Pu electronic structure since, as we state in the Introduction, a mere 1 wt % of Ga stabilizes the fcc δ phase.[34] Several other elements are equally as effective in transforming the α phase into δ, including americium. Indeed, aging effects have been the subject of much conjecture. While we do not claim to have carried out a comprehensive study of aging, we have obtained PES spectra in both freshly prepared as well as 10-yr-old α-Pu where about 0.1% of the atoms have undergone a disintegration. These spectra are shown in Fig. 8 using both He I and He II radiation. Very little difference is observed with 21.2 eV photons where the $6d$ electron cross section is emphasized, while at 40.8 eV photon energy, where the $5f$ cross section becomes significant, the near-E_F peak of the fresh material shows a substantial intensity increase (but no change in width) vs the aged material. This suggests that (a) impurities affect primarily the $5f$ orbitals, (b) again this is evidence of a significant $6d$ admixture in the near-E_F peak, which to a first approximation is unaffected by impurities, and (c) impurities are not the source of the narrow, near-E_F peak which confounds comparison to the LDA.

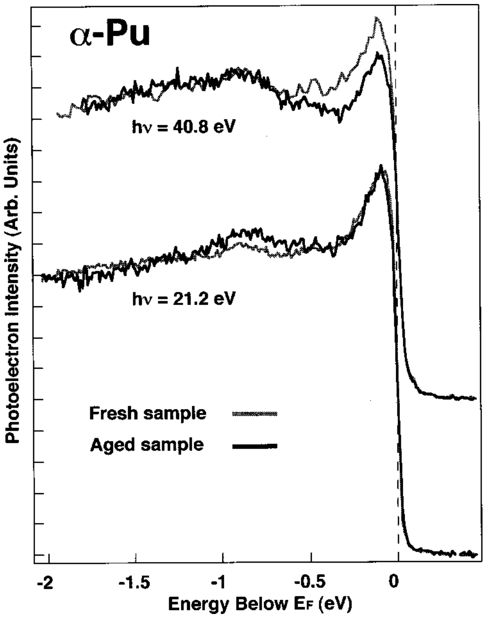

FIG. 8. Effects of aging in α-Pu. Note that effects are observed only at 40.8 eV where the PES spectrum is sensitive to $5f$ intensity. This may be indicative of greater bonding of $5f$ states vs $6d$ states to accumulated impurities.

We did not measure $4f$ core-level spectra, but these have been used in the past[33] to verify the more localized nature of δ-Pu vs α-Pu. The basic differences between $4f$ core-level satellites in α-Pu and δ-Pu have been discussed before in terms of the SIM model[32] and should be addressed here. In both cases the $4f$ core-level spectra show a metalliclike state at about 421 eV binding energy and a broad satellite at about 423 eV. The satellite is much more prominent in δ-Pu and has thus been modeled[32] within the SIM interpretation. However, the satellite is not negligible even in α-Pu, where a SIM interpretation seems less justified (e.g., the magnetic susceptibility shows temperature-independent paramagnetism[5]). Perhaps the existence and strength of the satellites is primarily related to the narrowness of the renormalized $5f$ band structure (and hence to their screening capability) and is not necessarily a significant indicator of SIM-like correlation effects.

We note for completeness that there exists previous takeoff-angle-dependent work[28] that suggests a possible surface reconstruction to a δ phase in α-Pu. It is indeed puzzling that the theoretical calculation for δ-Pu comes closer to reproducing the experimental data, including the sharp feature at (actually, only near) the Fermi edge, than it does for the presumably more strongly hybridized α-Pu. In addition, work has been reported[35] on thin films of α-Pu which is interpreted as indicating a δ-like surface reconstruction based on the spectral similarity argument. However, in that work the escape depth was not varied via a photon-energy dependence. By contrast, our measurements of the α-Pu spectra as a function of escape depth yield nearly invariant results. At least in bulk polycrystalline Pu a surface reconstruction seems unlikely, particulary not in the presence of pervasive surface oxygen.

IV. CONCLUSIONS

The similarity between α- and δ-Pu spectra raises suspicion of a surface reconstruction in α-Pu to a δ-like surface. However, the invariance with escape depth in both α-and δ-Pu indicates that the spectra presented here (as well as higher-energy PES data[36]) are representative of the bulk electronic structure. All data are consistent with the concept of much narrower itinerant states in δ-Pu vs α-Pu. Indeed, bulk property data for δ-Pu suggest heavy-fermion-like behavior. The narrow bands are nearly reproduced within a constrained LDA band calculation (with some residual discrepancies) suggesting no apparent need for further band narrowing arising from correlation effects. Temperature-independent PES data for both α- and δ-Pu support the notion that the SIM is not applicable. The nearly identical nature of the spectra (only subtle differences exist) below the Fermi energy is surprising and not reproduced in calculations. Further, the relatively narrow and intense α-Pu feature at E_F is likewise not reproduced. The introduction of correlation effects via periodic models such as the periodic Anderson model or renormalized band model may yet be necessary to understand this electronic structure, but the constrained LDA result for δ-Pu provides a reasonable starting point.

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