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Plasmas with index of refraction greater than one

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Abstract

Over the last decade, X-ray lasers in the wavelength range14 – 47 nm have been used to do interferometry of plasmas. Just as for optical interferometry of plasmas, the experimental analysis assumed that the index of refraction is due only to the free electrons. This makes the index of refraction less then one. Recent experiments in Al plasmas have observed fringe lines bend the wrong way as though the electron density is negative. We show how the bound electrons can dominate the index of refraction in many plasmas and make the index greater than one or enhance the index such that one would greatly overestimate the density of the plasma using interferometry.

OCIS number(s): 340.7450 X-ray interferometry, 120.3180 Interferometry, 120.5710 Refraction, 140.7240 UV, XUV, and X-ray lasers

The basic assumption since the first x-ray laser interferometers using the 15.5 nm Ne-like Y laser at the NOVA facility at Lawrence Livermore National Laboratory (LLNL) a decade ago is that the index of refraction of a plasma can be calculated from the free electron density.[1] The same assumption has been used for optical interferometry of plasmas for many decades.[2] Recent experiments[3] at the COMET laser facility at LLNL using the 14.7 nm Ni-like Pd laser and at the Advanced Photon Research Center[4] at JAERI using the 13.9 nm Ni-like Ag laser have observed anomalous behavior of fringe lines in interferometer experiments of Al plasmas where the fringe lines bend the wrong way, indicating the electron density is negative, or the index of refraction is greater than one. Experiments at both facilities observed the plasmas at late time when the plasmas are cooling and may only be a few times ionized.

In this work we discuss how the index of refraction for Al is far more complicated than generally assumed because there are significant contributions to the index from the continuum and line structure of the bound electrons that can dominate the free electron contribution and even change the sign of the gradient of the index with respect to the plasma density. We will also show how Al is not unique and how the bound electrons can have significant contributions for any plasma under the right conditions.

The usual formula for the index of refraction of a plasma due to free electrons is $n = (1 – N_{elec} / N_{crit})^{1/2}$ where N_{elec} is the electron density of the plasma and N_{crit} is the plasma critical density at a particular wavelength. For neutral materials the complex index of refraction is typically written as $n = 1 – \delta – i\beta$ where β is related to the absorption coefficient and $1 – \delta$ is the real part of the index of refraction that is relevant for interferometry.[5] The coefficient $\delta = 0.5 \times f1 \times (N_{ion} / N_{crit})$ where N_{ion} is the ion density and N_{crit} is the plasma critical density. For an ionized plasma the contribution to the index of refraction due to free electrons uses the same formula for δ but f1 is replaced by the number of free electrons per ion. This assumes the first order Taylor expansion of the square root. For most neutral materials, $f1 > 0$, but for neutral Al at 14.7 nm and 13.9 nm, f1 = -1.4 and –0.8, respectively. This raises the interesting question of whether Al is

unique and what the index is for singly ionized Al and other nearby charge states at these wavelengths.

The other important question is whether the X-ray laser can propagate through the partially ionized Al plasma. We used a Hartree-Slater code to calculate the absorption coefficient f2 for singly and double ionized Al and compared this with the published data[5] for neutral Al in Fig. 1. Keep in mind that $\beta = 0.5 \times f2 \times (N_{ion} / N_{crit})$. If one considers a plasma with an ion density of 10^{20} cm^{-3}, then the mean free path for 14.7 nm (84.46 eV) X-rays is only 24 µm for neutral Al but jumps to 512 and 1060 µm for singly and doubly ionized Al. This is because the absorption edge for Al moves from 73.2 eV for neutral Al to 91.7 and 105.4 eV for singly and doubly ionized Al. The result is that the X-ray laser goes from being highly absorbed by neutral Al to being on the less absorptive side of the L-edge for the partially ionized Al. This suggests that neutral Al does not likely play a large role in the anomaly because the plasma would be very absorptive. Typical interferometer experiments use Al plasmas with lengths of 1000 µm.[3,4] The question then becomes what is the index of refraction for the partially ionized Al.

To address this we calculated the continuum absorption cross sections for each ionization stage of Al using our Hartree-Slater code. The energies were adjusted to make sure the L3 edges for the 2p electrons agree with the experimentally measured edges for singly, doubly, and triply ionized Al. We then added the absorption for the lines below the L3 edges to the continuum absorption. For Al (+1) and Al (+2) we used the measured line positions and oscillator strengths from Refs. 6 and 7. For Al (+3) the n=2 to n=3 and 2p to 4s line positions and line strengths were used from Ref. 8. From the absorption coefficient f2, the real part of the index, f1, is then derived using the Kramers-Kronig dispersion relation. This part of the index is the bound electron contribution. We then add the number of free electrons to determine a total value for f1. Ref. 5 has a description of the Kramers-Kronig dispersion relation. Table 1 shows our best calculation of the partial components and total f1 value for each ionization stage of Al for a 14.7 nm X-ray.

From Table 1 one can see immediately that there can be large contributions to f1 from both the absorption lines and from the continuum absorption edges. When f1 is negative the index of refraction is greater than one. If one looks at the ratio of f1-total to the number of free electrons this is the same as the ratio of the measured electron density to the actual electron density when we use the usual analysis that only considers the free electron contribution. One can greatly underestimate the electron density for low ionization states. In fact one needs to reach Al (+7) before the contribution from the bound electrons becomes less than a 10% effect.

To understand how important the line contribution can be let us look at doubly ionized Al (+2), which has a large line contribution. Looking at the data in Ref. 6, there is a strong line measured at 89.89 eV with oscillator strength, f_{osc}, of 0.377. The incremental contribution to f1 from the wings of a single line, $\Delta f1(E) = f_{osc} / [(E / E_0)^2 - 1]$, where E_0 is the energy of the line. For our X-ray laser at 14.7 nm (84.46 eV), $\Delta f1 = -3.22$. For the Ag laser at 13.9 nm (89.25 eV), $\Delta f1 = -26.6$. The value of $\Delta f1$ is very sensitive to the exact position of the line and its line width as you approach resonance.

Having shown that the bound electrons can have a significant contribution to the index of refraction for an Al plasma the question remains as to whether this issue is important in other materials that do not have neutral absorption edges near the X-ray laser energy. If one looks at the ionization energies across the periodic chart a typical material has its first ionization about 5 to 10 eV. As it continues to ionize there will almost always be an edge near the X-ray laser energy that needs to be considered.

For Ti we calculate f1 for Ti (+4), Ti (+12), Ti (+13) and compare these with neutral Ti (+0). For each ionization stage we calculated the photo-ionization cross-sections with our Hartree-Slater code. Using the Grant code[9] we then calculated oscillator strengths for the strongest lines and added these to the absorption cross section. We estimate the term f1 by using the Kramers-Kronig dispersion relation and then add the free electrons to calculate a total value for f1.

In Fig. 2 we compare our estimated f1 for four-times ionized Ti (+4) with neutral Ti (+0). Ti (+4) is interesting because it is a stable sequence with closed 3s and 3p sub-shells. It has an ionization potential of 98 eV. From Fig. 2 one sees that at 84.46 eV (14.7 nm), f1 = 10.3 for neutral Ti and still has a value of 11.0 for Ti (+4). For Ti (+4) we expect the total f1 = 4 from the free electrons so in an actual experiment we would overestimate the electron density by 11/4 or 175% when you take the ratio of these numbers. If we look at Ti (+12) where the ionization potential is 788 eV one expects all the absorption edges and lines to be at much higher energy than the X-ray laser energy and therefore the bound electrons should make a very small contribution. For this case we estimate the total f1 to be 12.2, which is very close to the value of 12 for just the free electrons. If we do the same calculation of f1 for Ti (+13) we discover that there are 2 weak 2s-2p lines at 96 and 102 eV that perturb the value of f1. At 14.7 nm or 84.46 eV the effect is small and we estimate the total f1 value to be 13.0, which is the same as the number of free electrons. However as you approach the resonances the effect from these small lines can be significant.

In order to better understand the physics of the Ni-like Pd X-ray laser, experiments have been done to look at the electron density of the Pd plasma prior to lasing. Taking ten-times ionized Pd (+10) as an example, we calculate the optical constants and compare with neutral Pd (+0) using the same procedure as described for Ti above. Pd (+10) has an ionization potential of 239 eV so we might expect a significant abundance of this ion for plasmas below 50 eV. At 14.7 nm (84.46 eV), neutral Pd has f1 = 22 while Pd (+10) has f1 = 43. In both cases we would greatly overestimate the electron density of the plasma using the free electron approximation. For the Pd (+10) one can see that the strong lines, especially the 4p – 4d line estimated to be at 75 eV with oscillator strength of 7, have a large contribution to f1. One needs to remember that this problem is very complex and these results are only approximate. For all these calculations we assume the plasma is only in the ground state of the particular ionic stage while a real plasma will have a distribution of population in excited states.

We have shown how the index of refraction at 14.7 nm in an Al plasma can be greater than one for a plasma that is a few times ionized. This explains why the fringe lines bent the opposite way that was expected in recent interferometry of Al plasmas.[3,4] We have also shown how the index of refraction from the free electrons can be greatly enhanced in other plasmas when the contribution from the bound electrons is included. This can have important consequences for plasma diagnostics such as interferometry and needs to be carefully considered when analyzing experiments. This enhanced index of refraction will also affect the refraction and propagation of x-rays through a plasma as it effectively changes the critical density in the plasma.

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Table 1. Partial contributions and total value of optical constant f1 at photon energy 84.46 eV (14.7 nm) for different isoelectronic stages of Al.

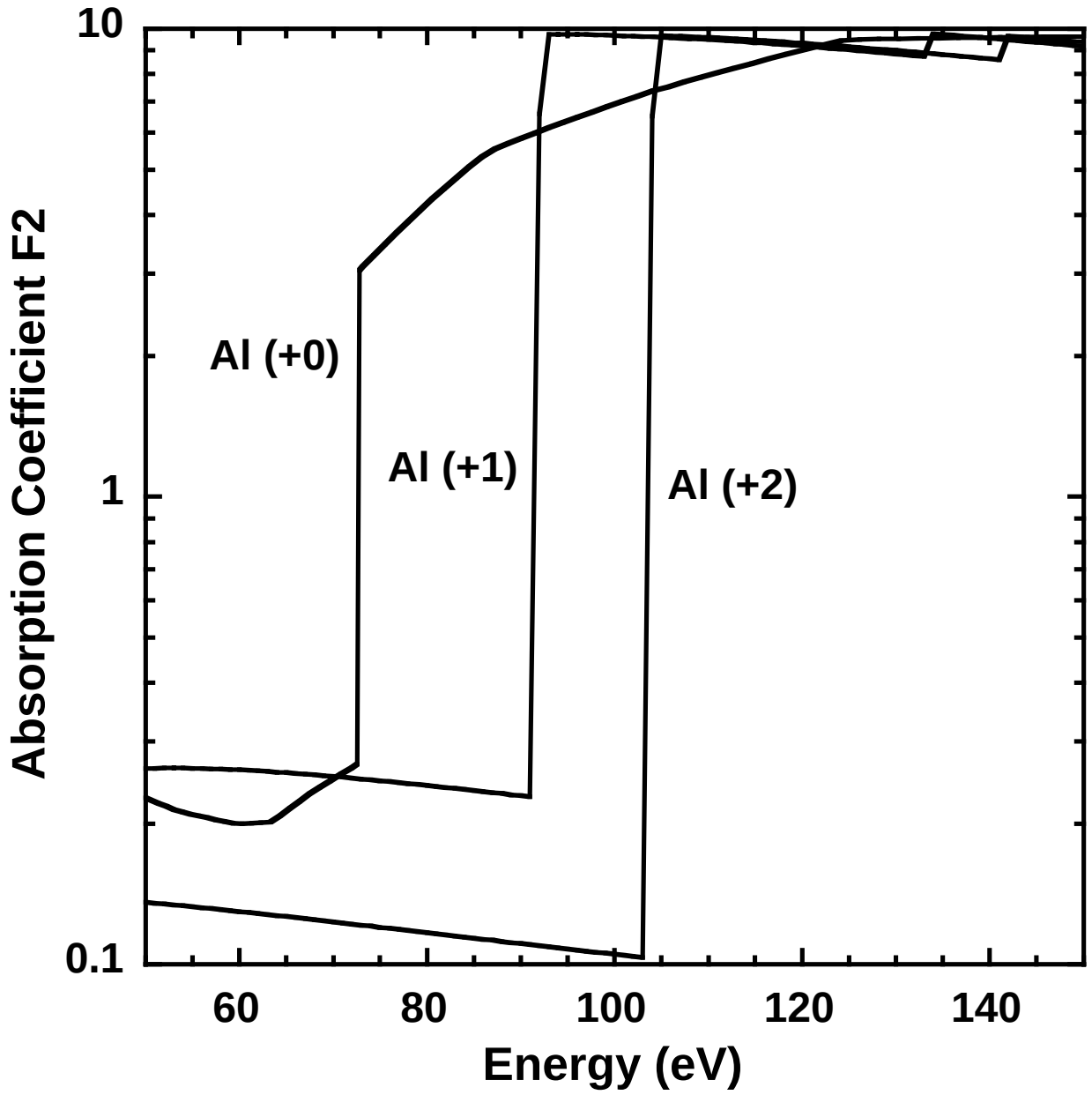
Ion	L3 edge (eV)	Line	Continuum	Free	F1-total
+0	73.1	2.61	-3.46	0.00	-0.85
+1	92.36	-0.70	-4.48	1.00	-4.19
+2	105.4	-2.91	-2.63	2.00	-3.54
+3	119.99	-3.16	-1.64	3.00	-1.80
+4	153.71	-2.49	-0.66	4.00	0.84
+5	190.47	-1.17	-0.29	5.00	3.54
+6	241.43	-0.57	-0.13	6.00	5.30
+7	284.59	-0.20	-0.06	7.00	6.73
+8	330.21	0.03	-0.03	8.00	8.00
+9	398.57	0.19	-0.02	9.00	9.18

Figure Captions

Fig. 1. Absorption coefficient f2 vs photon energy for neutral Al(+0), singly ionized Al(+1), and doubly ionized Al(+2).

Fig. 2. Optical constant f1 versus photon energy for neutral Ti(+0) (dotted) and four-times ionized Ti(+4) (solid). This includes the contribution from the free electrons.

Fig. 3. Optical constant f1 versus photon energy for neutral Pd(+0) (dotted) and ten-times ionized Pd(+10) (solid). This includes the contribution from the free electrons.



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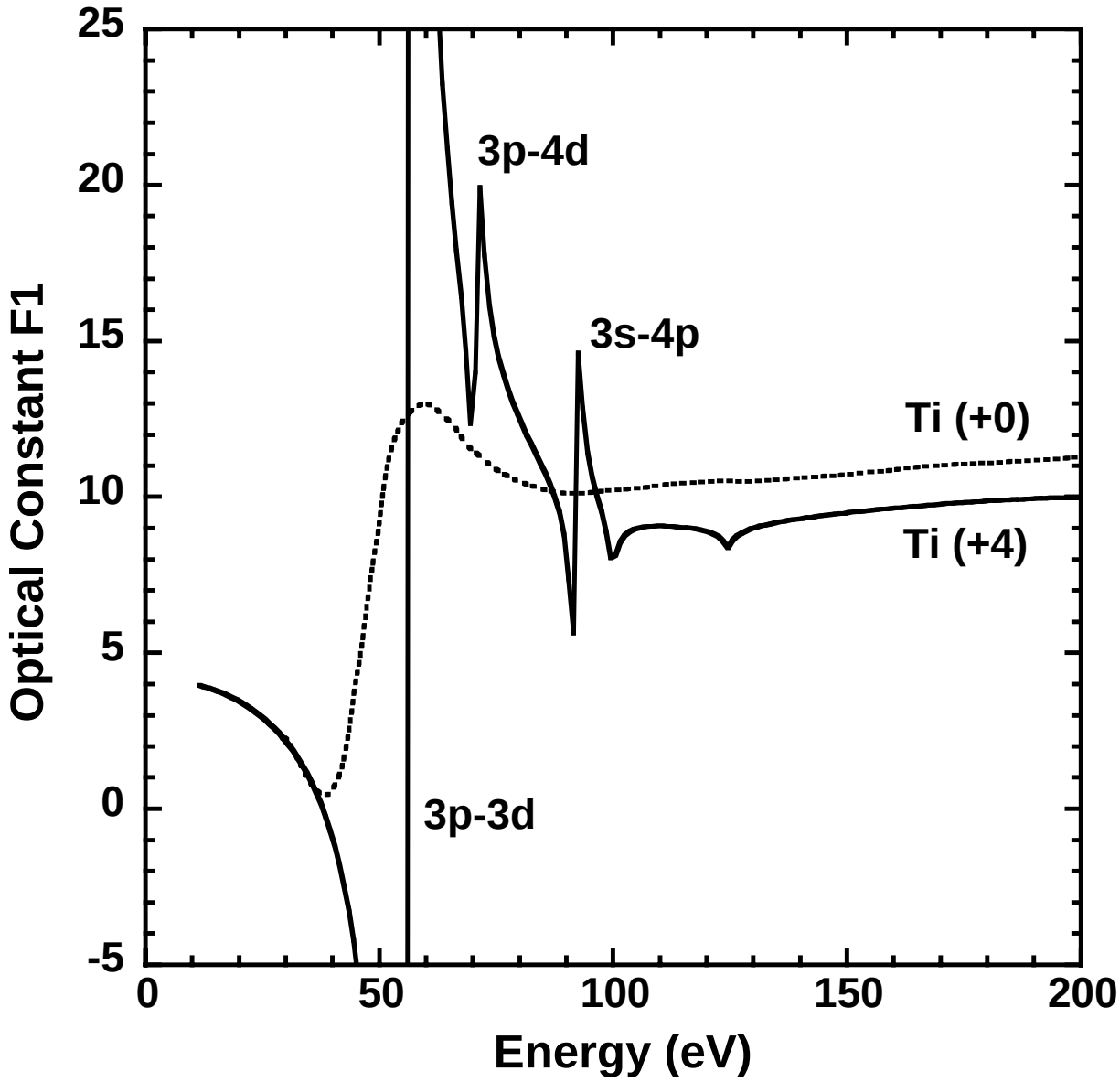

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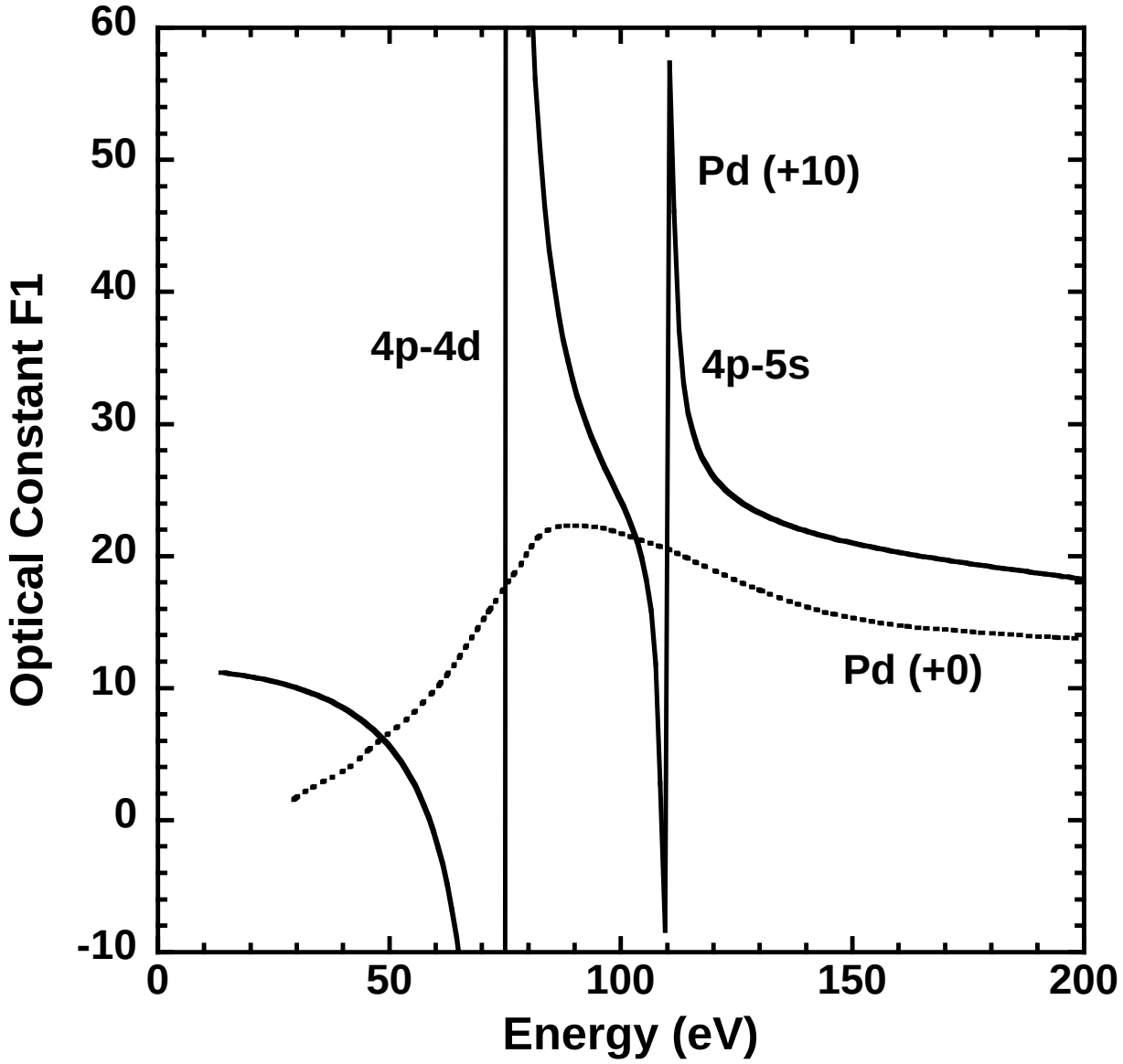

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