First-principles Computational Material Research

Lecture Notes

Calculations on Optical Properties

DAY 2

Lecturer : T. C. Leung（ 梁贊全 ）

Department of Physics

National Chung Cheng University

VASP Home Page

http://cms.mpi.univie.ac.at/vasp/vasp/vasp.html

Optics in VASP

Dielectric matrix in the random phase approximation (RPA)

Fouier transform

$$\vec{E}(\vec{r},t)=\int\frac{d^3q}{(2\pi)^3}\int\frac{d\omega}{2\pi}\vec{E}(q,\omega)e^{i(\vec{q}\cdot\vec{r}-\omega t)}$$

$$\vec{E}(\vec{q},\omega)=\int d^3r\int dt\vec{E}(\vec{r},t)e^{-i(\vec{q}\cdot\vec{r}-\omega t)}$$

$$\int d^3r\int_{-\infty}^{\infty}dt\left(\frac{\partial\vec{E}}{\partial t}\right)e^{-i(\vec{q}\cdot\vec{r}-\omega t)}=\int d^3re^{-i(\vec{q}\cdot\vec{r})}\left[e^{i\omega t}E\Big|_{-\infty}^{\infty}-i\omega\int_{-\infty}^{\infty}dt\vec{E}e^{i(\omega t)}\right]$$

$$\frac{\partial\vec{E}}{\partial t}\xrightarrow{F.T.}-i\omega\vec{E}(q,\omega)$$

$$\vec{\nabla}\cdot\vec{E}\xrightarrow{F.T.}i\vec{q}\cdot\vec{E}(q,\omega)\xrightarrow{L-field}i|q||E(q,\omega)|$$

$$\nabla^2\phi\xrightarrow{F.T.}-q^2\phi(q,\omega)$$

$$\vec{\nabla}\cdot\vec{E}(\vec{r},t)=4\pi\rho(\vec{r},t)=-e4\pi n(\vec{r},t)$$

$$iqE(\vec{q},\omega)=-e4\pi n(\vec{q},\omega)$$

$$\nabla^2V(\vec{r},t)=\nabla^2(-e)\phi(\vec{r},t)=4\pi e^2n(\vec{r},t)$$

$$V(\vec{q},\omega)=\frac{4\pi e^2}{|q|^2}n(\vec{q},\omega)$$

Dielectric function

$$\vec{E}_{ext}(\vec{r},\omega) = \int d^3r'\varepsilon(\vec{r},\vec{r}',\omega)\vec{E}_{tot}(\vec{r}',\omega)$$

note $\vec{D} = \varepsilon\vec{E}$

$$\vec{E}_{tot}(\vec{r},\omega) = \int d^3r'\varepsilon^{-1}(\vec{r},\vec{r}',\omega)\vec{E}_{ext}(\vec{r}',\omega)$$

$$\Rightarrow \quad \boxed{\vec{E}_{tot}(\vec{q},\omega) = \int d^3q'\varepsilon^{-1}(\vec{q}',\omega)\vec{E}_{ext}(\vec{q}',\omega)}$$

$$\varepsilon(\vec{r},\vec{r}',\omega) = \int \frac{d^3q'}{(2\pi)^3}\int \frac{d^3q''}{(2\pi)^3} e^{i\vec{q}'\cdot\vec{r}}\ \varepsilon(\vec{q}',\vec{q}'',\omega)\, e^{-i\vec{q}''\cdot\vec{r}'}$$

$$\varepsilon(\vec{r}+\vec{R},\vec{r}'+\vec{R},\omega) = \varepsilon(\vec{r},\vec{r}',\omega) \quad \Rightarrow \quad \vec{q}'-\vec{q}'' = \vec{G}$$

$$\Rightarrow \quad \vec{q}' = \vec{q}+\vec{G}; \qquad \vec{q}'' = \vec{q}+\vec{G}'$$

$$\vec{E}_{total}(\vec{q}+\vec{G},\omega) = \sum_{\vec{G}'}\varepsilon^{-1}(\vec{q}+\vec{G},\vec{q}+\vec{G}',\omega)\vec{E}_{ext}(\vec{q}+\vec{G}',\omega)$$

$$\Rightarrow \quad \boxed{\vec{E}_{tot}(\vec{q}+\vec{G},\omega) = \sum_{\vec{G}'}\varepsilon^{-1}_{\vec{G}\vec{G}'}(\vec{q},\omega)\vec{E}_{ext}(\vec{q}+\vec{G}',\omega)}$$

$$\vec{E}_{ext}(\vec{q}+\vec{G},\omega)=\sum_{\vec{G}'}\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)\vec{E}_{tot}(\vec{q}+\vec{G}',\omega)$$

$$iqE(\vec{q},\omega)=-e4\pi n(\vec{q},\omega)$$

$$V(\vec{q},\omega)=\frac{4\pi e^2}{|q|^2}n(\vec{q},\omega)$$

$$\frac{n_{ext}(\vec{q}+\vec{G},\omega)}{|\vec{q}+\vec{G}|}=\sum_{\vec{G}'}\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)\frac{n_{tot}(\vec{q}+\vec{G}',\omega)}{|\vec{q}+\vec{G}'|}$$

$$n_{ext}(\vec{q}+\vec{G},\omega)=\sum_{\vec{G}'}|\vec{q}+\vec{G}|\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)|\vec{q}+\vec{G}'|\frac{n_{tot}(\vec{q}+\vec{G}',\omega)}{(\vec{q}+\vec{G}')^2}$$

$$-n_{ind}(\vec{q}+\vec{G},\omega)=\sum_{\vec{G}'}|\vec{q}+\vec{G}||\vec{q}+\vec{G}'|\left[\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)-\delta_{\vec{G}\vec{G}'}\right]\frac{n_{tot}(\vec{q}+\vec{G}',\omega)}{(\vec{q}+\vec{G}')^2}$$

$$-n_{ind}(\vec{q}+\vec{G},\omega)=\sum_{\vec{G}'}|\vec{q}+\vec{G}||\vec{q}+\vec{G}'|\left[\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)-\delta_{\vec{G}\vec{G}'}\right]\frac{V_{tot}(\vec{q}+\vec{G}',\omega)}{4\pi}$$

$$-\frac{\partial n_{ind}(\vec{q}+\vec{G},\omega)}{\partial V_{tot}(\vec{q}+\vec{G}'',\omega)}=\left[\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)-\delta_{\vec{G}\vec{G}'}\right]\frac{|\vec{q}+\vec{G}||\vec{q}+\vec{G}''|}{4\pi}$$

$$\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)=\delta_{\vec{G}\vec{G}'}-\frac{4\pi e^2}{|\vec{q}+\vec{G}||\vec{q}+\vec{G}'|}\frac{\partial n_{ind}(\vec{q}+\vec{G},\omega)}{\partial V_{tot}(\vec{q}+\vec{G}',\omega)}=\delta_{\vec{G}\vec{G}'}-\nu_{\vec{G}\vec{G}'}P_{\vec{G}\vec{G}'}(\vec{q},\omega)$$

RPA approximation

$$P_{\vec{G}\vec{G}'}(\vec{q},\omega)=\chi^{KS}_{\vec{G}\vec{G}'}(\vec{q},\omega)$$

$$\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)=\delta_{\vec{G}\vec{G}'}-\frac{4\pi e^2}{|\vec{q}+\vec{G}||\vec{q}+\vec{G}'|}\frac{\delta n_{ind}(\vec{q}+\vec{G},\omega)}{\delta V_{tot}(\vec{q}+\vec{G}',\omega)}=\delta_{\vec{G}\vec{G}'}-v_{\vec{G}\vec{G}'}\chi^{KS}_{\vec{G}\vec{G}'}(\vec{q},\omega)$$

$$\chi^{KS}_{\vec{G}\vec{G}'}(\vec{q},\omega)==-\frac{1}{\Omega}\sum_{n\vec{k}}^{all}\sum_{m\vec{k}+\vec{q}}^{all}2f_{n\vec{k}}(1-f_{m\vec{k}+\vec{q}})$$ (Appendix 1-1)

$$\times\left[\frac{\langle\psi_{m\vec{k}+\vec{q}}|e^{i(\vec{q}+\vec{G})\cdot\vec{r}}|\psi_{n\vec{k}}\rangle\langle\psi_{n\vec{k}}|e^{-i(\vec{q}+\vec{G}')\cdot\vec{r}}|\psi_{m\vec{k}+\vec{q}}\rangle}{\varepsilon_{m\vec{k}+\vec{q}}-\varepsilon_{n\vec{k}}-\varpi}+\frac{\langle\psi_{n\vec{k}}|e^{i(\vec{q}+\vec{G})\cdot\vec{r}}|\psi_{m\vec{k}+\vec{q}}\rangle\langle\psi_{m\vec{k}+\vec{q}}|e^{-i(\vec{q}+\vec{G}')\cdot\vec{r}}|\psi_{n\vec{k}}\rangle}{\varepsilon_{m\vec{k}+\vec{q}}-\varepsilon_{n\vec{k}}+\varpi}\right]$$

$$\langle\psi_{m\vec{k}'}|e^{i(\vec{q}+\vec{G})\cdot\vec{r}}|\psi_{n\vec{k}}\rangle=\langle u_{m\vec{k}+\vec{q}}|e^{i\vec{G}\cdot\vec{r}}|u_{n\vec{k}}\rangle\xrightarrow{\vec{G}=0}\langle\psi_{m\vec{k}+\vec{q}}|e^{i\vec{q}\cdot\vec{r}}|\psi_{n\vec{k}}\rangle=\langle u_{m\vec{k}+\vec{q}}|u_{n\vec{k}}\rangle$$

If the off-diagonal elements of the dielectric matrix (local field effects) are neglected.

$$\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)=\varepsilon_{\vec{G}\vec{G}'}(\vec{q},\omega)\delta_{\vec{G}\vec{G}'}\Rightarrow\varepsilon_{mac}(\vec{q},\omega)=\varepsilon_{00}(\vec{q},\omega)$$ (Appendix 2)

$$\varepsilon_{mac}(\hat{q},\omega) = \lim_{\vec{q}\to 0} \varepsilon_{00}(\vec{q},\omega) = 1 - \lim_{\vec{q}\to 0} \frac{4\pi e^2}{q^2} \chi^{KS}_{0,0}(\vec{q},\omega)$$

$$\varepsilon_{mac}(\hat{q},\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$$

$$\varepsilon_\infty^{(2)}(\hat{\mathbf{q}},\omega) = \frac{4\pi^2 e^2}{\Omega} \lim_{\mathbf{q}\to 0} \frac{1}{|\mathbf{q}|^2} \sum_{c,v,\mathbf{k}} 2w_{\mathbf{k}} \delta(\epsilon_{c\mathbf{k}+\mathbf{q}} - \epsilon_{v\mathbf{k}} - \omega) \times |\langle u_{c\mathbf{k}+\mathbf{q}} | u_{v\mathbf{k}} \rangle|^2 \qquad (n \neq m)$$

$$\varepsilon_\infty(\hat{\mathbf{q}},\omega) = \lim_{\mathbf{q}\to 0} \varepsilon_\infty(\mathbf{q},\omega) = \sum_{\alpha,\beta} \hat{\mathbf{q}}_\alpha \varepsilon_{\alpha\beta}(\omega) \hat{\mathbf{q}}_\beta$$

$$\varepsilon^{(2)}_{\alpha\beta} = \frac{4\pi^2 e^2}{\Omega} \sum_{\mathbf{k}} \sum_{\mathrm{vc}} 2w_{\mathbf{k}} \delta(\varepsilon_{\mathrm{c},\mathbf{k}} - \varepsilon_{\mathrm{v},\mathbf{k}} - \omega) \times \langle \boldsymbol{\beta}_{\alpha,\mathrm{c},\mathrm{k}} | \tilde{\mathrm{u}}_{\mathrm{v},\mathbf{k}} \rangle \langle \tilde{\mathrm{u}}_{\mathrm{v},\mathbf{k}} | \boldsymbol{\beta}_{\beta,\mathrm{c},\mathrm{k}} \rangle$$

$$\omega_p^2 = \frac{4\pi e^2}{m^2} \sum_{nk} \left| \vec{\varepsilon} \cdot \overrightarrow{M}_{nn}(\vec{k}) \right|^2 \left(-\frac{\partial f_0}{\partial \varepsilon} \right)_{\varepsilon_{nk}} D_n(\hbar\omega) \qquad (n = m)$$

$$\varepsilon^{(1)}_{\alpha\beta}(\omega) = 1 + \frac{2}{\pi} \mathrm{P} \int_0^\infty \frac{\varepsilon^{(2)}_{\alpha\beta}(\omega')\omega'}{\omega'^2 - \omega^2} d\omega'$$

Phys. Rev. B 73, 45112 (2006)

Comp. Phys. Commun. 175, 1 (2006)

How to evalute $\lim_{\mathbf{q}\to 0}\langle \psi_{n'\mathbf{k}+\mathbf{q}}|e^{i\mathbf{q}\mathbf{r}}|\psi_{n\mathbf{k}}\rangle$

$$e^{i\mathbf{q}(\mathbf{r}-\mathbf{R}_i)} = 1 + i\mathbf{q}(\mathbf{r}-\mathbf{R}_i) + o(\mathbf{q}^2)$$

$$\lim_{\mathbf{q}\to 0}\langle \psi_{n'\mathbf{k}+\mathbf{q}}|e^{i\mathbf{q}\mathbf{r}}|\psi_{n\mathbf{k}}\rangle = \langle \tilde{u}_{n'\mathbf{k}+\mathbf{q}}|\left(1 + \sum_{ij} |\tilde{p}_{i\mathbf{k}+\mathbf{q}}\rangle Q_{ij}\langle \tilde{p}_{j\mathbf{k}}|\right)|\tilde{u}_{n\mathbf{k}}\rangle$$

$$+ i\mathbf{q}\sum_{ij} \langle \tilde{u}_{n'\mathbf{k}+\mathbf{q}}|\tilde{p}_{i\mathbf{k}+\mathbf{q}}\rangle \vec{\tau}_{ij}\langle \tilde{p}_{j\mathbf{k}}|\tilde{u}_{n\mathbf{k}}\rangle,$$

$$Q_{ij} = \int_{\Omega_{\mathrm{PAW}}} [\phi_i(\mathbf{r})\phi_j(\mathbf{r}) - \tilde{\phi}_i(\mathbf{r})\tilde{\phi}_j(\mathbf{r})]d^3\mathbf{r}$$

$$\vec{\tau}_{ij} = \int_{\Omega_{\mathrm{PAW}}} (\mathbf{r}-\mathbf{R}_i)[\phi_i(\mathbf{r})\phi_j(\mathbf{r}) - \tilde{\phi}_i(\mathbf{r})\tilde{\phi}_j(\mathbf{r})]d^3\mathbf{r}$$

Basics of the PAW formalism

$$|\psi_{n\mathbf{k}}\rangle = |\tilde{\psi}_{n\mathbf{k}}\rangle + \sum_i (|\phi_i\rangle - |\tilde{\phi}_i\rangle)\langle \tilde{p}_i|\tilde{\psi}_{n\mathbf{k}}\rangle$$

$$\langle \tilde{p}_i|\tilde{\phi}_j\rangle = \delta_{ij}$$

$$\langle \mathbf{r}|\tilde{p}_i\rangle = \tilde{p}_i(\mathbf{r}-\mathbf{R}_i)$$

$$|\tilde{\psi}_{n\mathbf{k}}\rangle = e^{i\mathbf{k}\mathbf{r}}|\tilde{u}_{n\mathbf{k}}\rangle$$

$$|\tilde{p}_{i\mathbf{k}}\rangle = e^{-i\mathbf{k}(\mathbf{r}-\mathbf{R}_i)}|\tilde{p}_i\rangle$$

$$\langle \tilde{p}_i|\tilde{\psi}_{n\mathbf{k}}\rangle = e^{i\mathbf{k}\mathbf{R}_i}\langle \tilde{p}_{i\mathbf{k}}|\tilde{u}_{n\mathbf{k}}\rangle$$

$$\tilde{u}_{n\mathbf{k}+\mathbf{q}} = \tilde{u}_{n\mathbf{k}} + \mathbf{q}\nabla_{\mathbf{k}}\tilde{u}_{n\mathbf{k}} + o(\mathbf{q}^2)$$

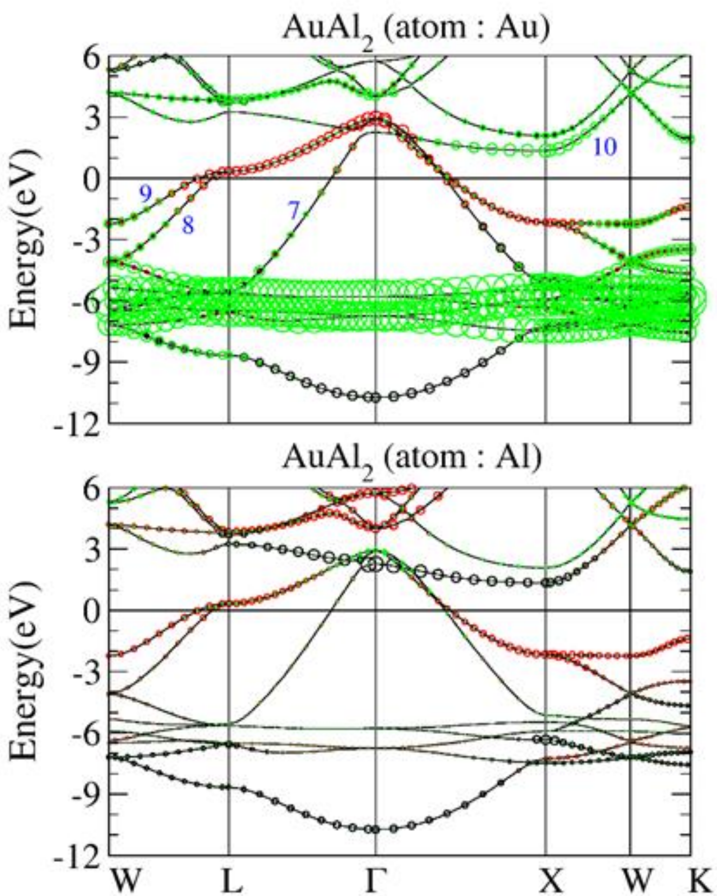
$$|\tilde{p}_{i\mathbf{k}+\mathbf{q}}\rangle = [1 - i\mathbf{q}(\mathbf{r}-\mathbf{R}_i)]|\tilde{p}_{i\mathbf{k}}\rangle + o(\mathbf{q}^2)$$

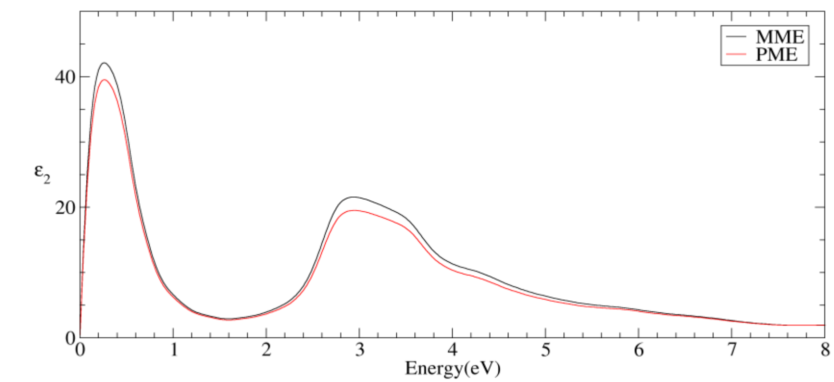
$$\lim_{\vec{q}\to 0}\left\langle \psi_{n'\vec{k}+\vec{q}}\middle| e^{i\vec{q}\cdot\vec{r}}\middle|\psi_{n'\vec{k}}\right\rangle = |\vec{q}|\left\langle \hat{q}\cdot\vec{\beta}_{n'\vec{k}}\middle|\tilde{u}_{n\vec{k}}\right\rangle$$

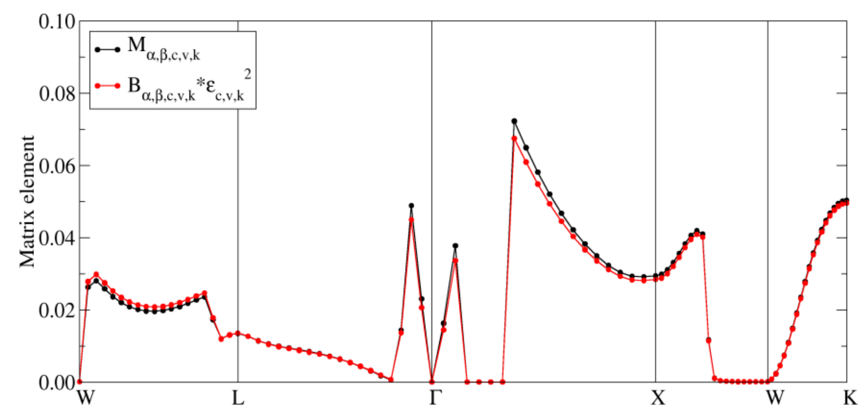
$$\left|\vec{\beta}_{n'\vec{k}}\right\rangle = \left(1+\sum_i\left|\tilde{p}_{i\vec{k}}\right\rangle Q_{ij}\left\langle\tilde{p}_{j\vec{k}}\right|\right)\left|\vec{\nabla}_{\vec{k}}\tilde{u}_{n\vec{k}}\right\rangle + i\left(\sum_{ij}\left|\tilde{p}_{i\vec{k}}\right\rangle Q_{ij}\left\langle\tilde{p}_{j\vec{k}}\right|\right)(\vec{r}_i-\vec{R}_i)\left|\tilde{u}_{n\vec{k}}\right\rangle - i\sum_{ij}\left|\tilde{p}_{i\vec{k}}\right\rangle\vec{\tau}_{ij}\left\langle\tilde{p}_{j\vec{k}}\middle|\tilde{u}_{n\vec{k}}\right\rangle$$

$$\varepsilon^{(2)}(\omega) = \frac{4\pi e^2}{m^2\omega^2}\sum_k 2w_k \sum_{v,c}\left|\vec{\varepsilon}\cdot\overrightarrow{M_{cv}}(\vec{k})\right|^2 \delta(\varepsilon_{ck}-\varepsilon_{vk}-\omega)$$

$$\varepsilon^{(2)}_{\alpha\beta} = \frac{4\pi^2 e^2}{\Omega}\sum_{\mathbf{k}}\sum_{vc} 2w_{\mathbf{k}}\delta(\varepsilon_{c,\mathbf{k}}-\varepsilon_{v,\mathbf{k}}-\omega)\times\langle\boldsymbol{\beta}_{\alpha,c,\mathbf{k}}|\tilde{u}_{v,\mathbf{k}}\rangle\langle\tilde{u}_{v,\mathbf{k}}|\boldsymbol{\beta}_{\beta,c,\mathbf{k}}\rangle$$







The matrix element for band 7 and 8 in $AuAl_2$. Notice that $\alpha=\beta=x$.

Drude model

$$\sigma = 1 + \frac{\sigma_0}{1 - i\omega\tau}, \quad \sigma_0 = \frac{ne^2\tau}{m}, \quad \omega_p^2 = \frac{4\pi ne^2}{m}$$

$$\varepsilon = 1 + \frac{4\pi i\sigma}{\omega} = 1 + \frac{4\pi i\sigma_0}{\omega(1 - i\omega\tau)} = 1 + \frac{i\omega_p^2\tau}{\omega(1 - i\omega\tau)} = 1 - \frac{\tau^2\omega_p^2}{(1+\omega^2\tau^2)} + \frac{i\tau\omega_p^2}{\omega(1+\omega^2\tau^2)}$$

$$\boxed{\begin{aligned}\varepsilon_1 &= 1 - \frac{\tau^2\omega_p^2}{(1+\omega^2\tau^2)} \\ \varepsilon_2 &= \frac{\tau\omega_p^2}{\omega(1+\omega^2\tau^2)}\end{aligned}} \xrightarrow{\tau = 1/\Gamma} \boxed{\begin{aligned}\varepsilon_1 &= 1 - \frac{\omega_p^2}{(\Gamma^2 + \omega^2)} \\ \varepsilon_2 &= \frac{\Gamma\omega_p^2}{\omega(\Gamma^2 + \omega^2)}\end{aligned}} \xrightarrow{\omega >> \Gamma} \boxed{\begin{aligned}\varepsilon_1 &= 1 - \frac{\omega_p^2}{\omega^2} \\ \varepsilon_2 &= \frac{\Gamma\omega_p}{\omega^3}\end{aligned}}$$

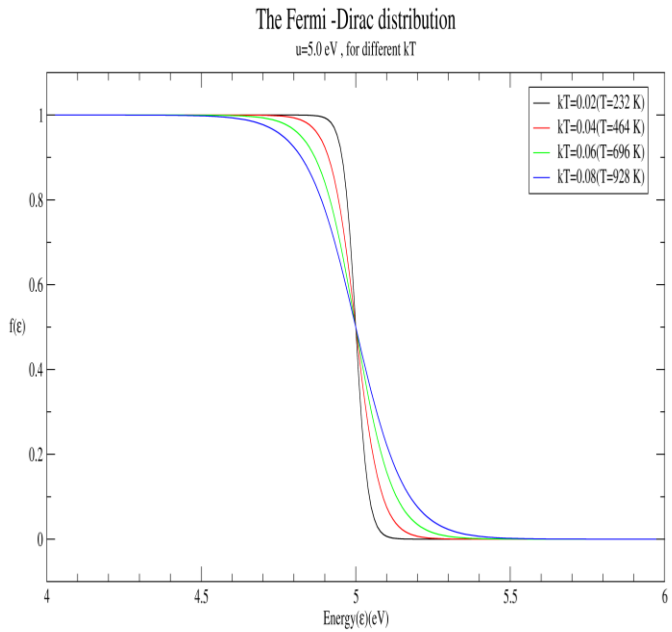
$$\omega_p^2 = \frac{4\pi e^2}{m^2} \sum_{nk} \left| \vec{\varepsilon} \cdot \overrightarrow{M_{nn}}(\vec{k}) \right|^2 \left(-\frac{\partial f_0}{\partial \varepsilon} \right)_{\varepsilon_{nk}} D_n(\omega)$$

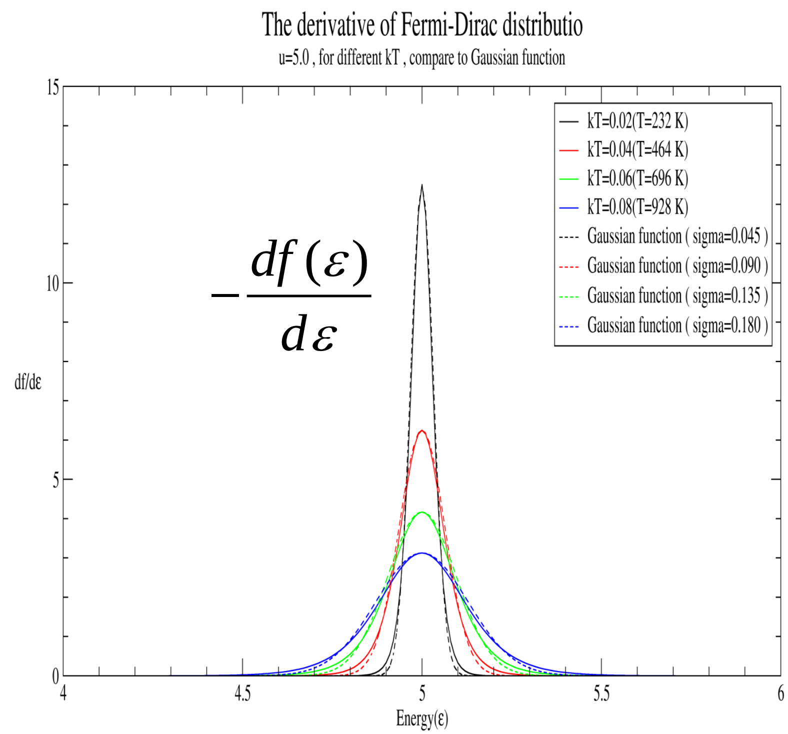
Energy loss function $= \mathrm{Im}\left(-\frac{1}{\varepsilon} \right) = \mathrm{Im}\left(-\frac{1}{\varepsilon_1 + i\varepsilon_2} \right) = \frac{\varepsilon_2}{\varepsilon_1^2 + \varepsilon_2^2}$

The fermi dirac function

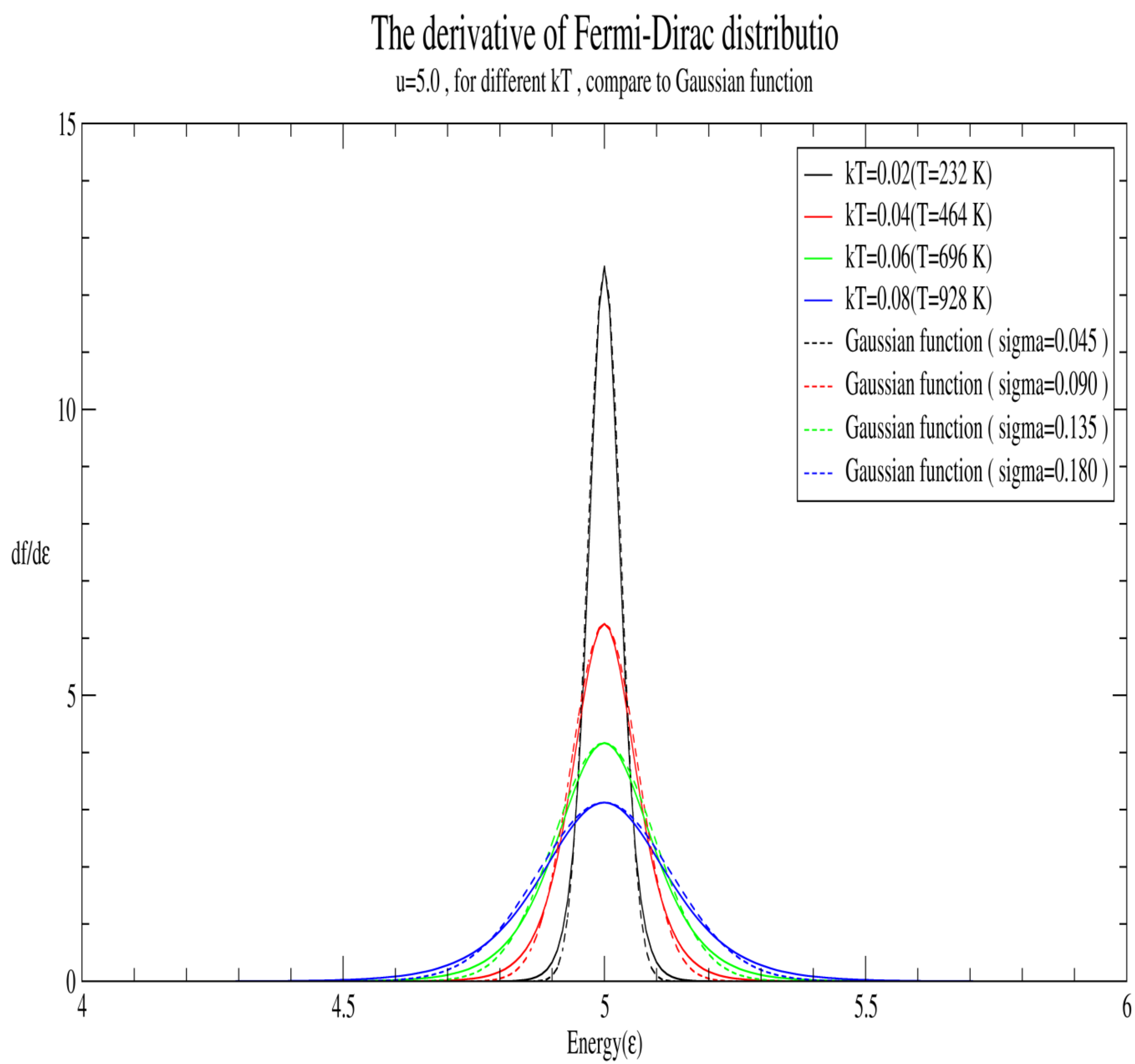
$$f(\varepsilon) = \frac{1}{1 + e^{(\varepsilon - \mu)/kT}}$$

At T = 0 $\qquad -\dfrac{df(\varepsilon)}{d\varepsilon} = \delta(\varepsilon - \mu)$

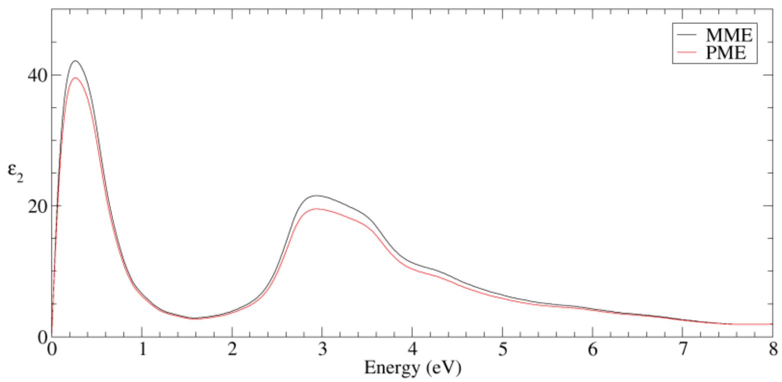




For small T, it behaves like Gaussian function $\qquad f(\varepsilon) = \dfrac{1}{\alpha\sqrt{\pi}} e^{-\left(\frac{\varepsilon - \mu}{\alpha}\right)^2}$

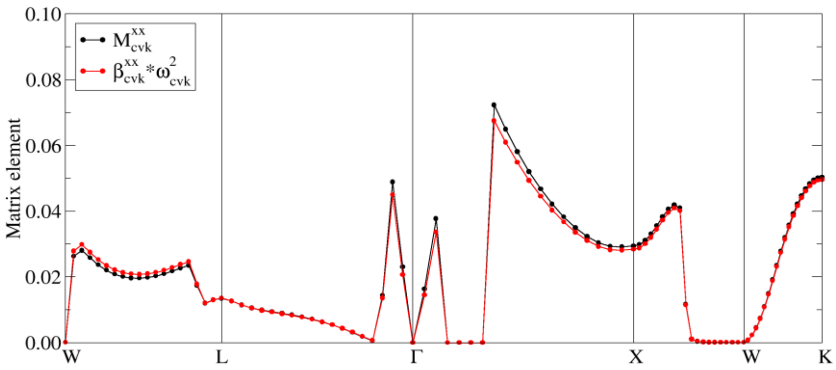

The derivative of Fermi-Dirac distributio
u=5.0 , for different kT , compare to Gaussian function
kT=0.02(T=232 K)
kT=0.04(T=464 K)
kT=0.06(T=696 K)
kT=0.08(T=928 K)
Gaussian function (sigma=0.045)
Gaussian function (sigma=0.090)
Gaussian function (sigma=0.135)
Gaussian function (sigma=0.180)
df/dε
Energy(ε)
0
5
10
15
4
4.5
5
5.5
6

Interband transition ε_2 of $AuAl_2$



$$\varepsilon_{xx}^{(2)}(\omega) = \frac{4\pi^2\hbar^2 e^2}{\Omega m^2} \sum_{k} \sum_{vc} 2w_{\mathbf{k}}\delta(\varepsilon_{c,\mathbf{k}}-\varepsilon_{v,\mathbf{k}}-\omega) \frac{\langle\psi_{c,k}|\hat{p}_x|\psi_{v,k}\rangle\langle\psi_{v,k}|\hat{p}_x|\psi_{c,k}\rangle}{\omega_{cvk}^2}$$

$$\varepsilon_{xx}^{(2)}(\omega) = \frac{4\pi^2 e^2}{\Omega} \sum_{k} \sum_{vc} 2w_{\mathbf{k}}\delta(\varepsilon_{c,\mathbf{k}}-\varepsilon_{v,\mathbf{k}}-\omega) \times \langle\boldsymbol{\beta}_{x,c,k}|\tilde{u}_{v,\mathbf{k}}\rangle\langle\tilde{u}_{v,\mathbf{k}}|\boldsymbol{\beta}_{x,c,k}\rangle$$



Matrix element for bands **7** and **8** of $AuAl_2$

$$M_{cvk}^{xx} \equiv \langle\Psi_{v,k}|\hat{p}_x|\Psi_{c,k}\rangle\langle\Psi_{c,k}|\hat{p}_x|\Psi_{v,k}\rangle$$

$$\beta_{cvk}^{xx} \equiv \langle\boldsymbol{\beta}_{x,c,k}|\tilde{u}_{v,k}\rangle\langle\tilde{u}_{v,k}|\boldsymbol{\beta}_{x,c,k}\rangle$$

$$M_{cvk}^{xx} \approx \beta_{cvk}^{xx} \times \omega_{cvk}^2$$

(suppose that $\hbar = m = 1$)

Electron energy-loss spectroscopy

$$L(\omega) = -\mathrm{Im}\left(\frac{1}{\varepsilon(\omega)}\right)$$

Enegy loss function

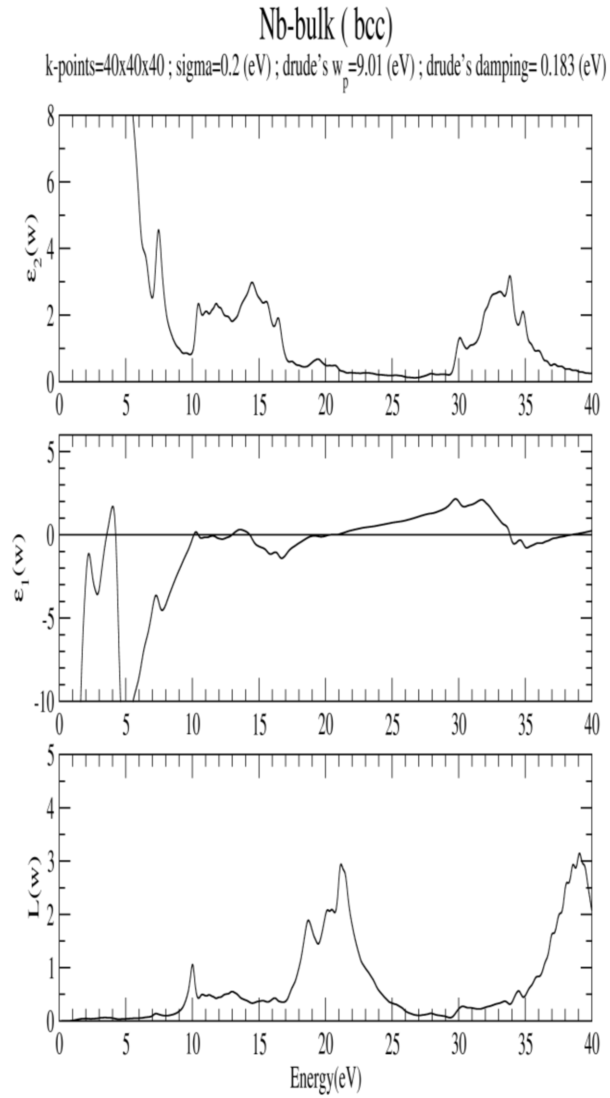
Complex refractive index

$$\tilde{n}(\omega) = n(\omega) + i\kappa(\omega)$$

$$\begin{cases} n(\omega) = \sqrt{\dfrac{\varepsilon_1(\omega)+|\varepsilon(\omega)|}{2}} \\ \kappa(\omega) = \sqrt{\dfrac{-\varepsilon_1(\omega)+|\varepsilon(\omega)|}{2}} \end{cases}$$

Reflectivity

$$R(\omega) = \frac{[1 - n(\omega)]^2 + \kappa^2(\omega)}{[1 + n(\omega)]^2 + \kappa^2(\omega)}$$



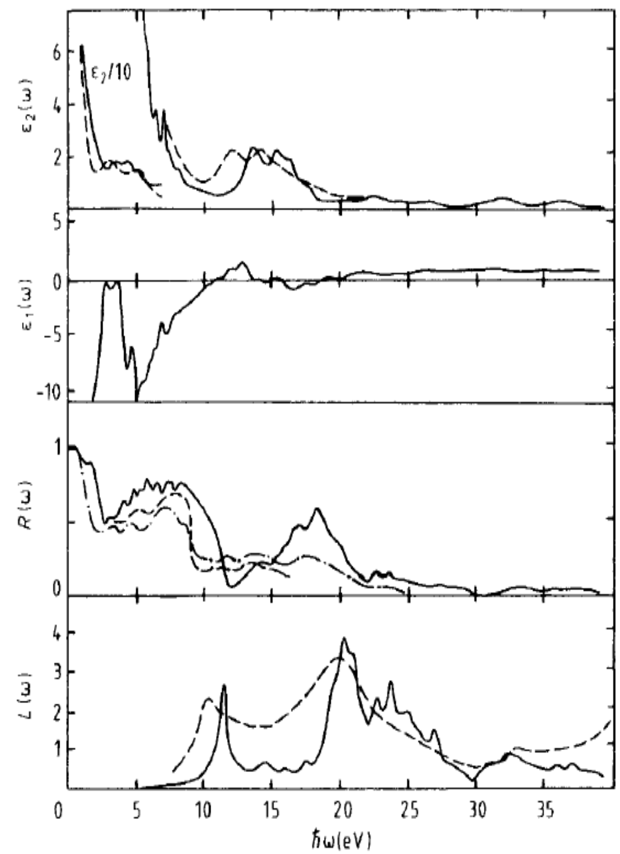
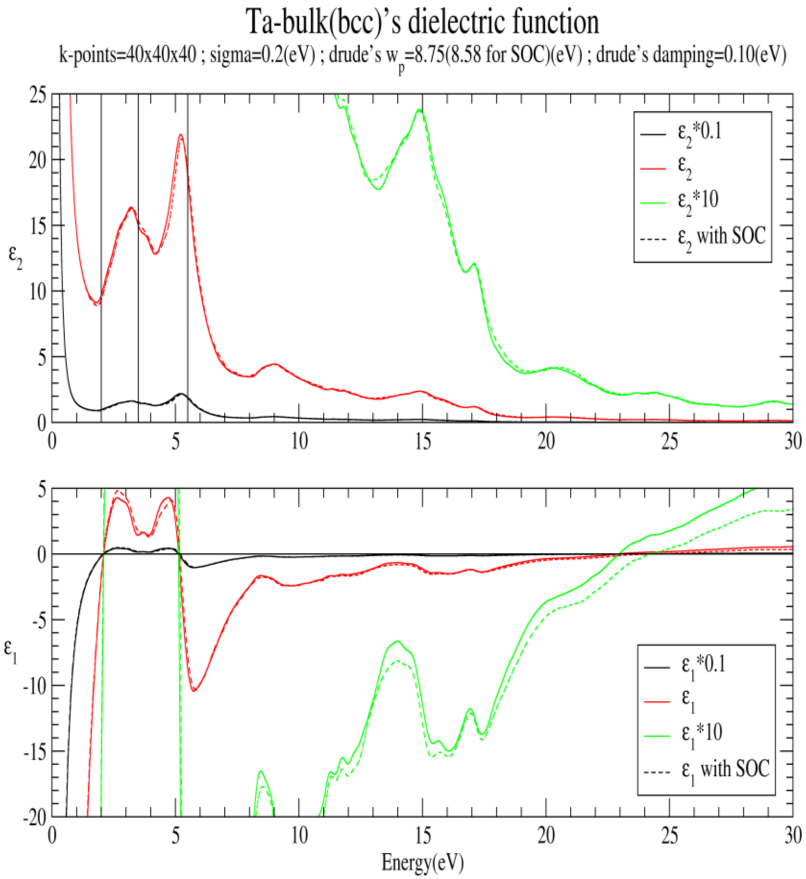


Figure 5. Optical properties of niobium: full curves, calculations; broken curves, measurements of $\varepsilon_2(\omega)$ (from Weaver *et al* 1973), $R(\omega)$ (from Vilesov *et al* 1967) and $L(\omega)$ (from Lynch and Swan 1968); chain curve, measurements of $R(\omega)$ (from Weaver *et al* 1973) (arbitrary units).

J.Phys.F:Met.Phys.18(1988) 833-849



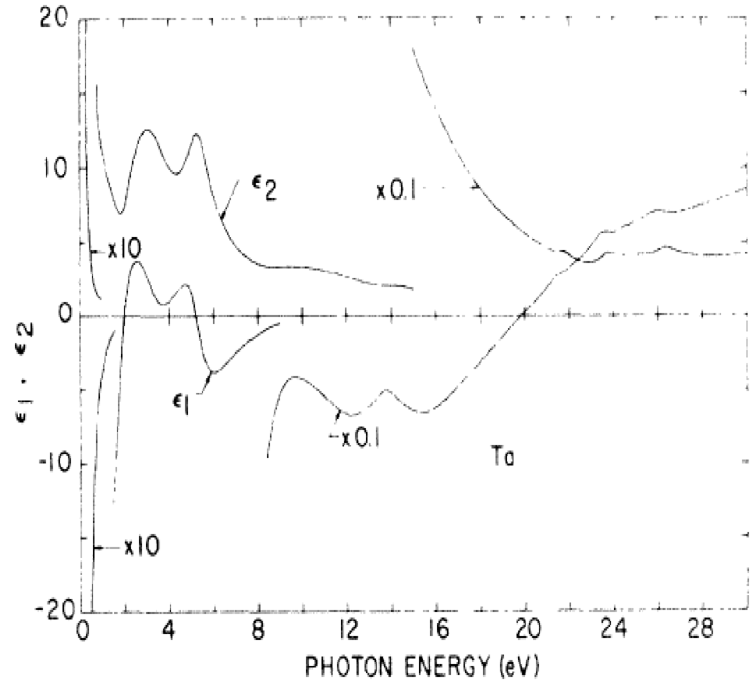
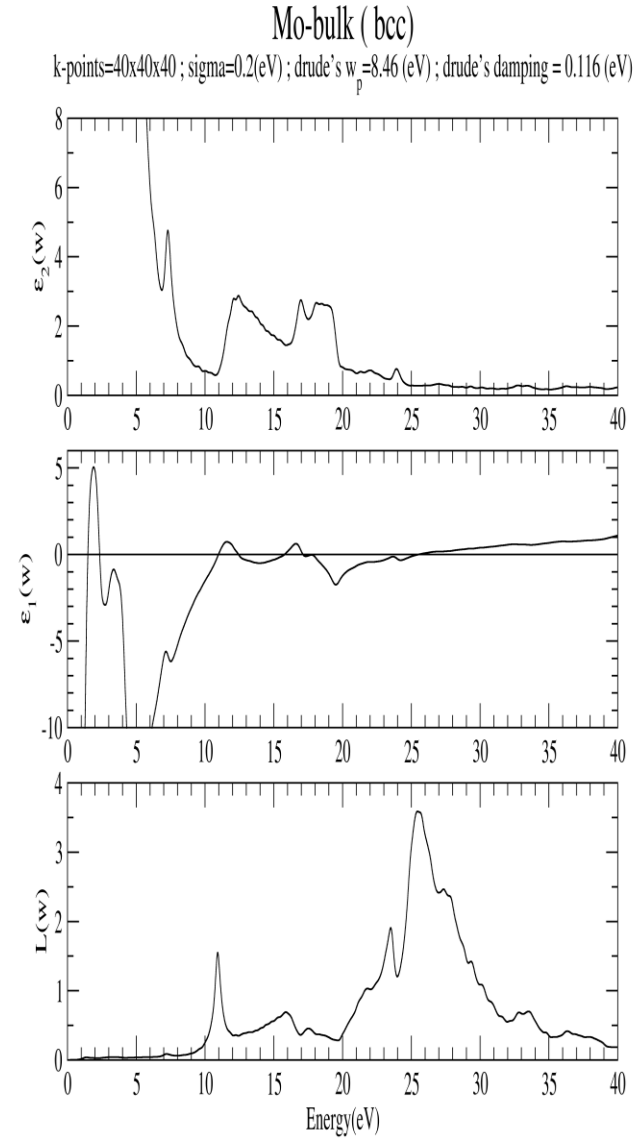


FIG. 6. Dielectric functions of Ta. The scales are shown expanded for clarity.

J.H.Weaver , Phys.Rev.B 10,501 (1974)



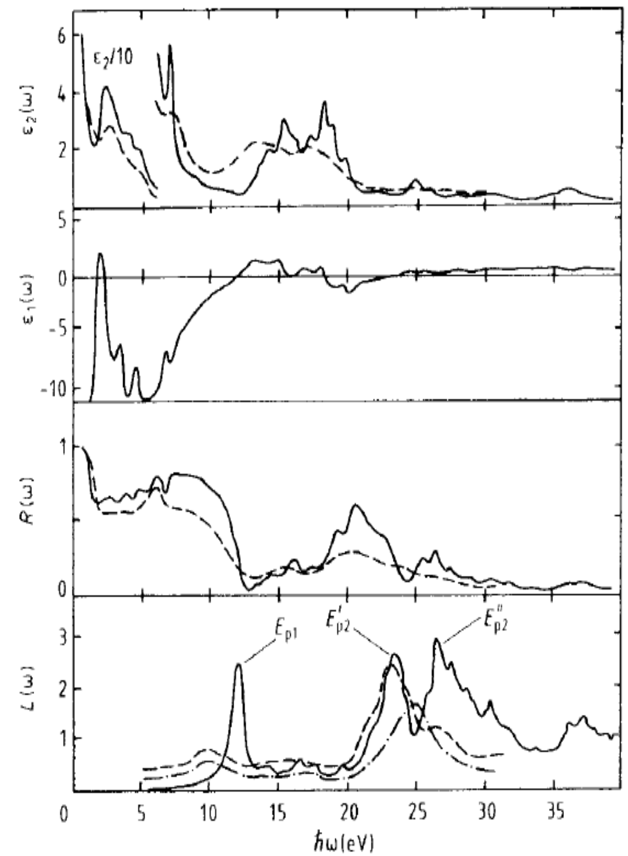
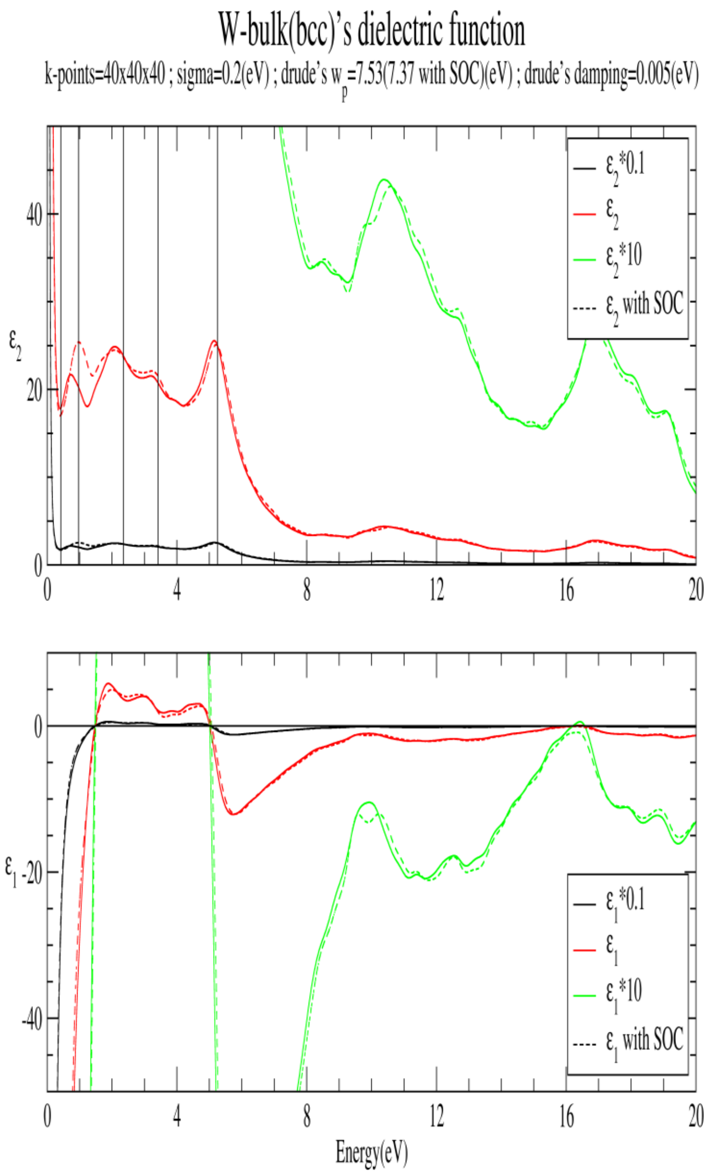


Figure 6. Optical properties of molybdenum: full curves, calculations; broken curves, measurements (from Mayevskii *et al* 1981); chain curve, measurements of $L(\omega)$ (from Weaver *et al* 1974) (arbitrary units).

J.Phys.F:Met.Phys.18(1988) 833-849



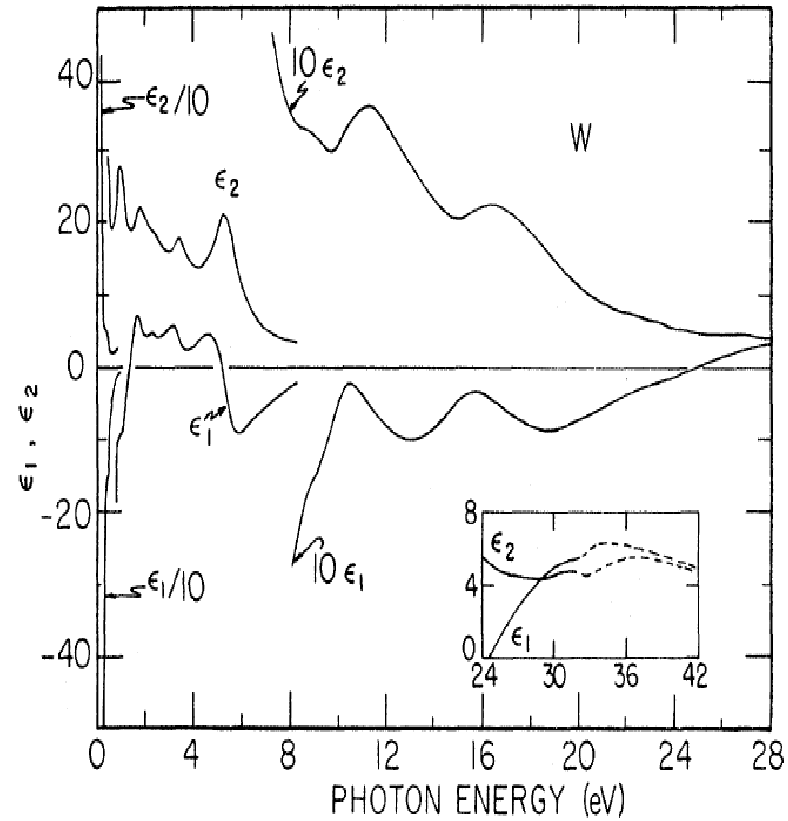
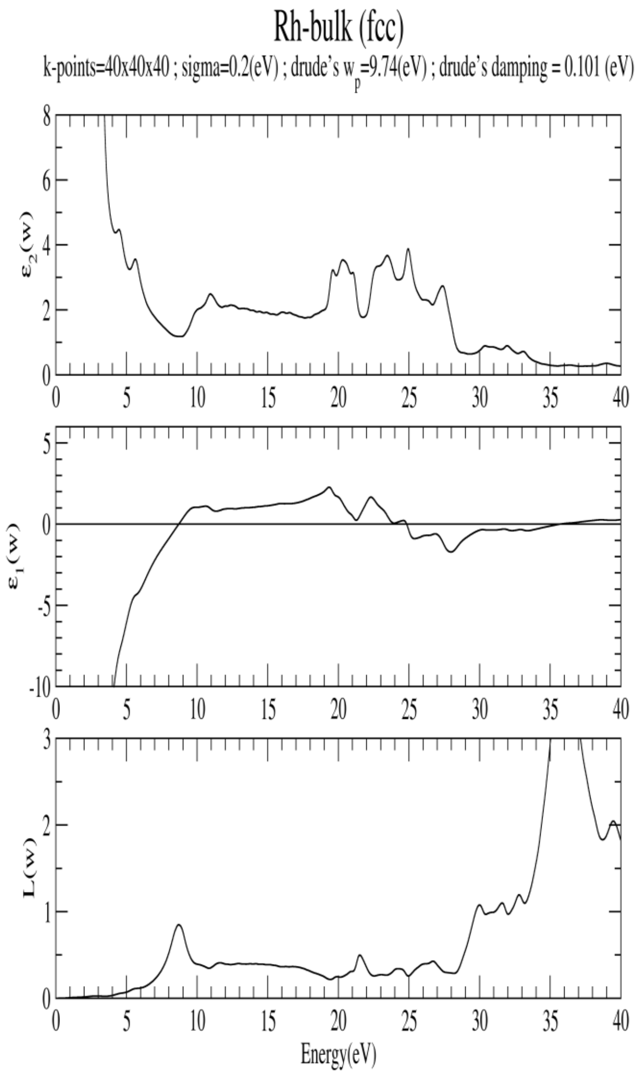


FIG. 3. Real and imaginary parts of the dielectric function of W. The high-energy region is shown in the insert.

J.H.Weaver Phy.Rev.B 12,1293(1975)



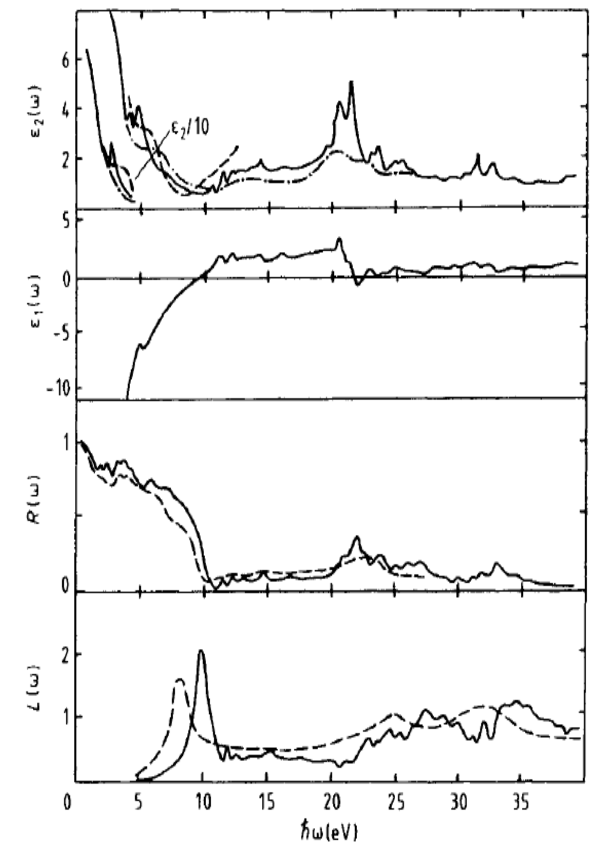
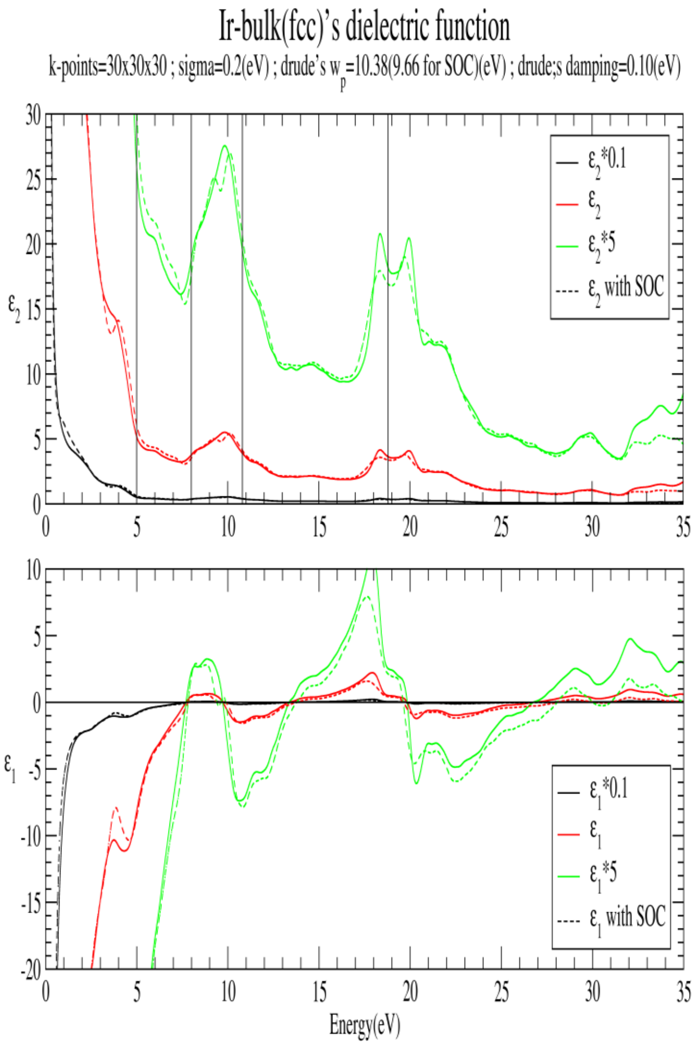


Figure 3. Optical properties of rhodium: full curves, calculations; broken curves, measurements of $\varepsilon_2(\omega)$ (from Pierce and Spicer 1973), $R(\omega)$ (from Seignac and Robin 1970) and $L(\omega)$ (from Lynch and Swan 1968); chain curve, measurements of $\varepsilon_2(\omega)$ (from Seignac and Robin 1970) (arbitrary units).

J.Phys.F:Met.Phys.18(1988) 833-849



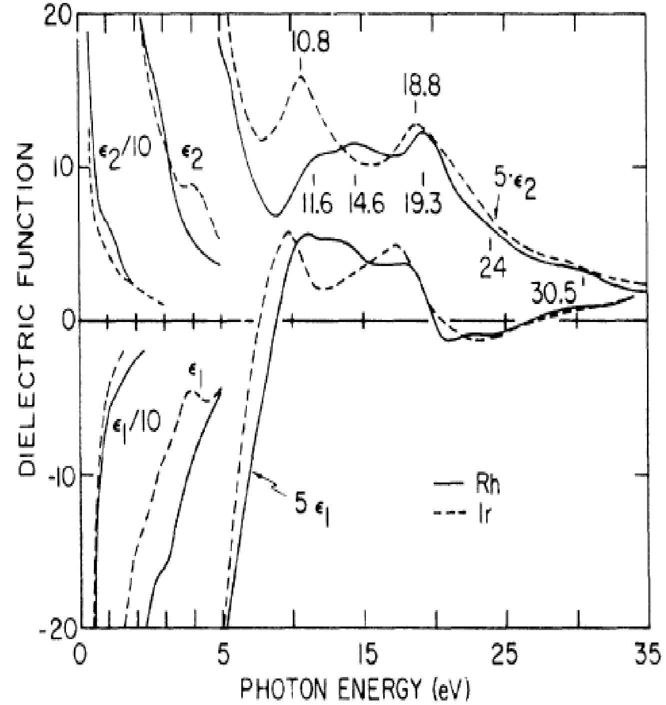
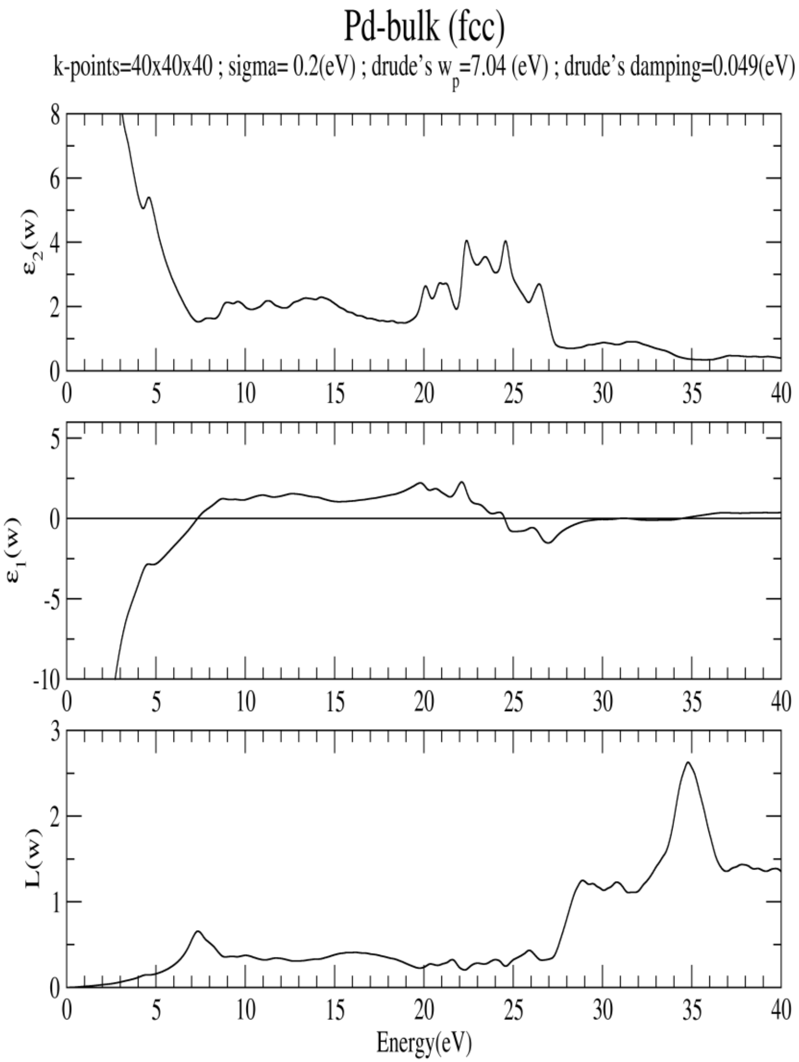


FIG. 3. Dielectric functions for Rh and Ir. The energy scale is linear, but changes at 5 eV. The low-energy structure is shown better in a plot of the conductivity (see Fig. 4).

J.H.Weaver , Phys.Rev.B 15,4115 (1977)



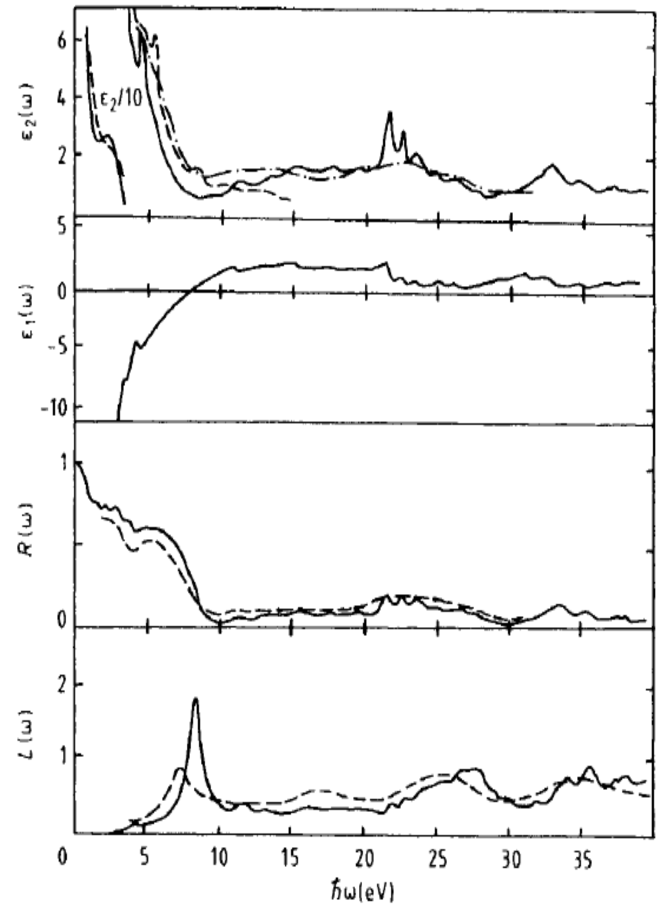
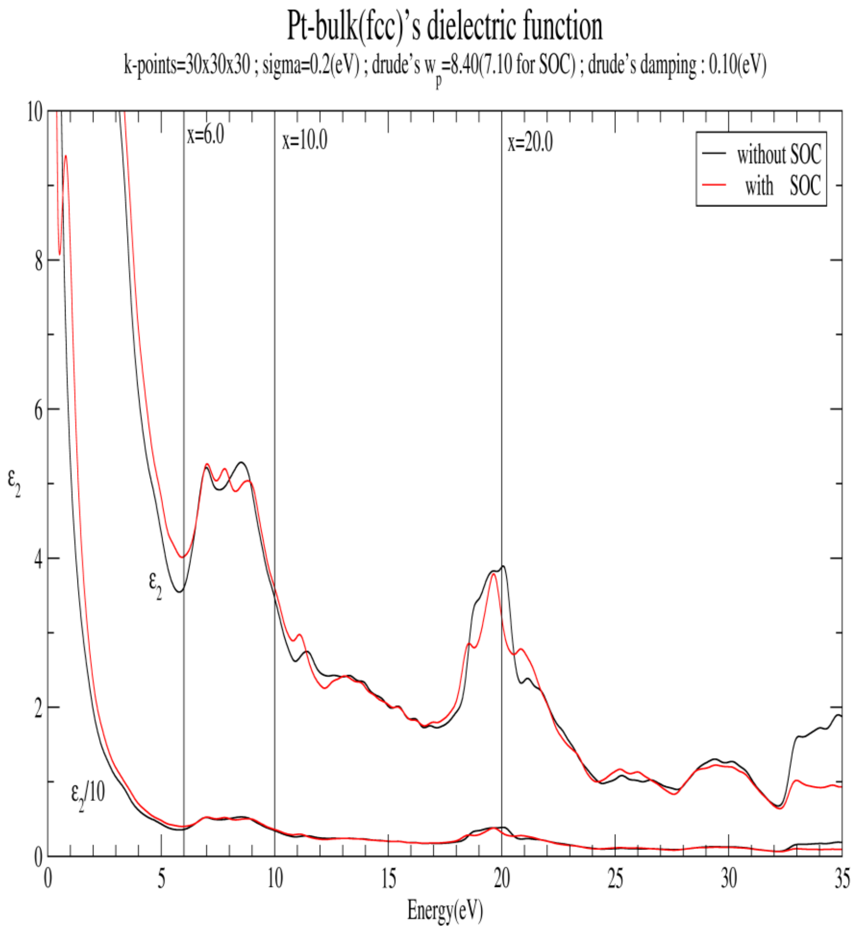


Figure 4. Optical properties of palladium: full curves, calculations; broken curves, measurements of $\varepsilon_2(\omega)$ (from Weaver 1973), $R(\omega)$ (from Vehse *et al* 1970) and $L(\omega)$ (from Daniels 1969); chain curve, measurements of $\varepsilon_2(\omega)$ (from Vehse *et al* 1970) (arbitrary units).

J.Phys.F:Met.Phys.18(1988) 833-849



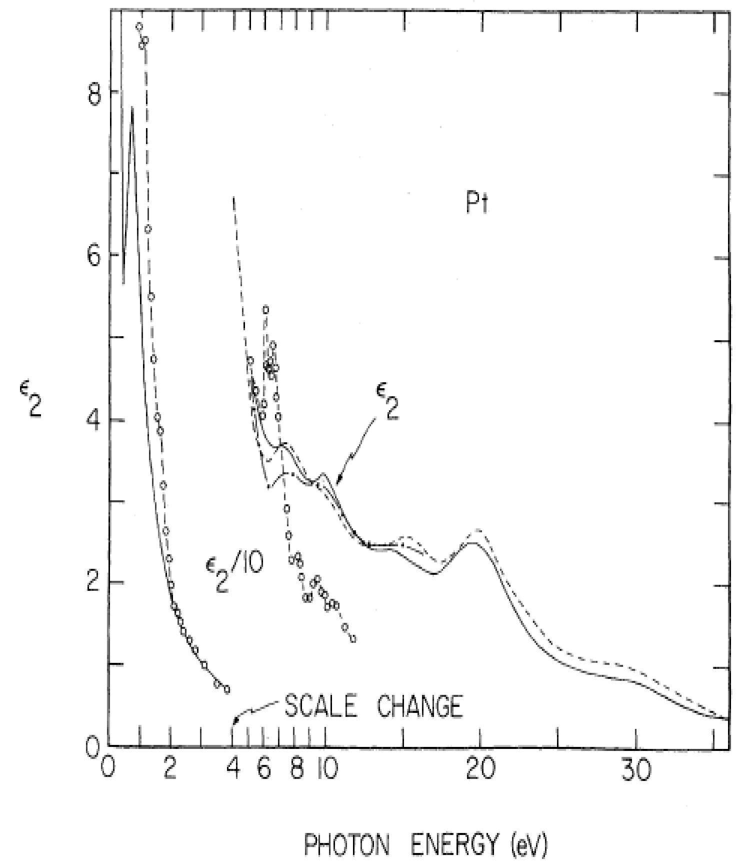
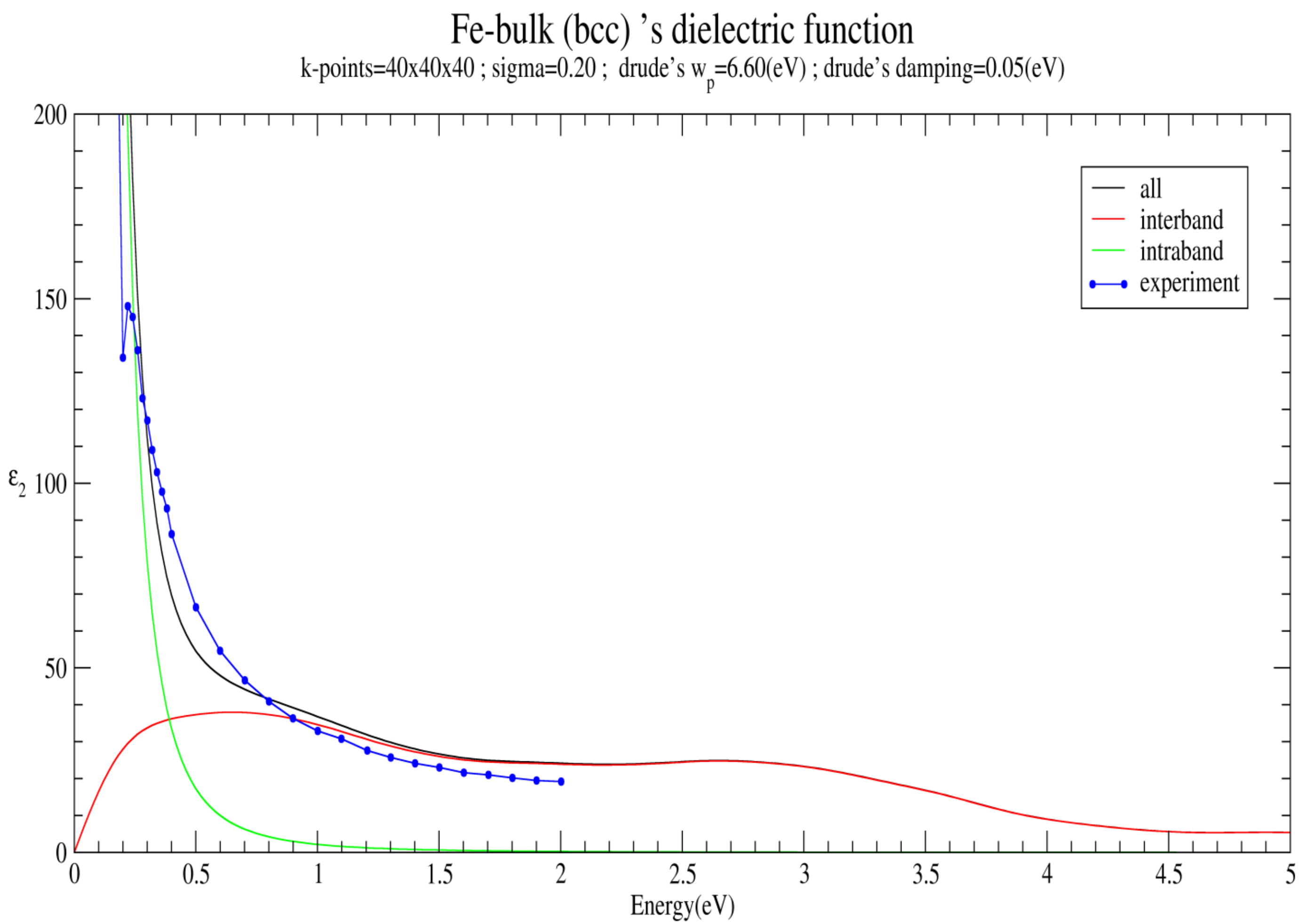


FIG. 8. ϵ_2 spectra of Pt. The calculated results are shown as open circles. The solid line represents the results obtained using the author's $R(E)$ data plus the high-energy $R(E)$ of Ref. 46. Experimental results are as follows: solid circles, Ref. 49; dashed line, Ref. 50. The energy scale is linear, but the scale doubles at 4 eV.

J.H.Weaver , Phys.Rev.B 11,1416 (1975)


Fe-bulk (bcc) 's dielectric function
k-points=40x40x40 ; sigma=0.20 ; drude's w_p=6.60(eV) ; drude's damping=0.05(eV)
all
interband
intraband
experiment
ε_2
0
50
100
150
200
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5
Energy(eV)

Beyond LDA

1. GW Approximation : include the self-energy

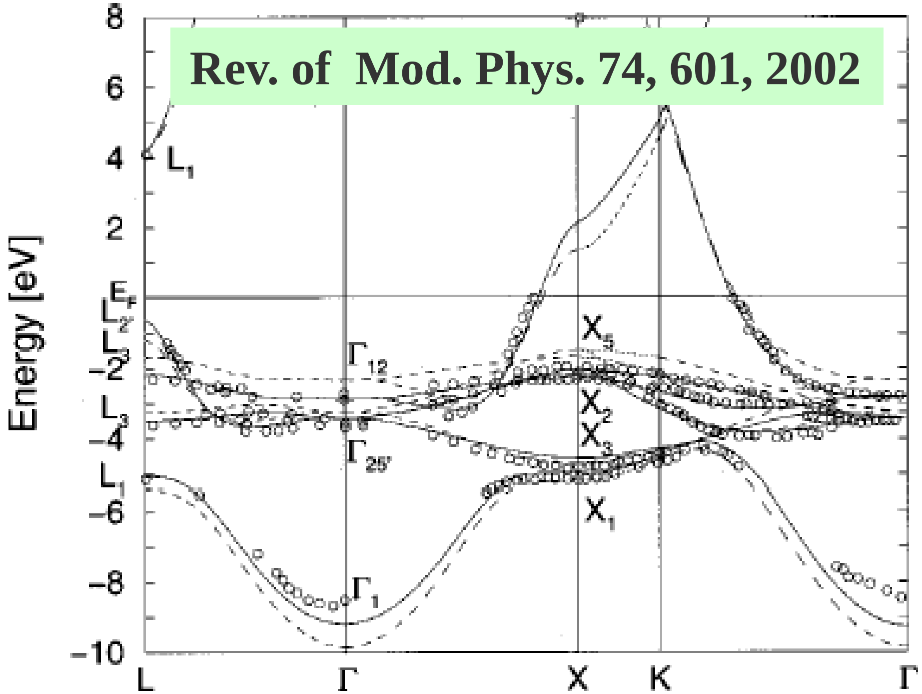


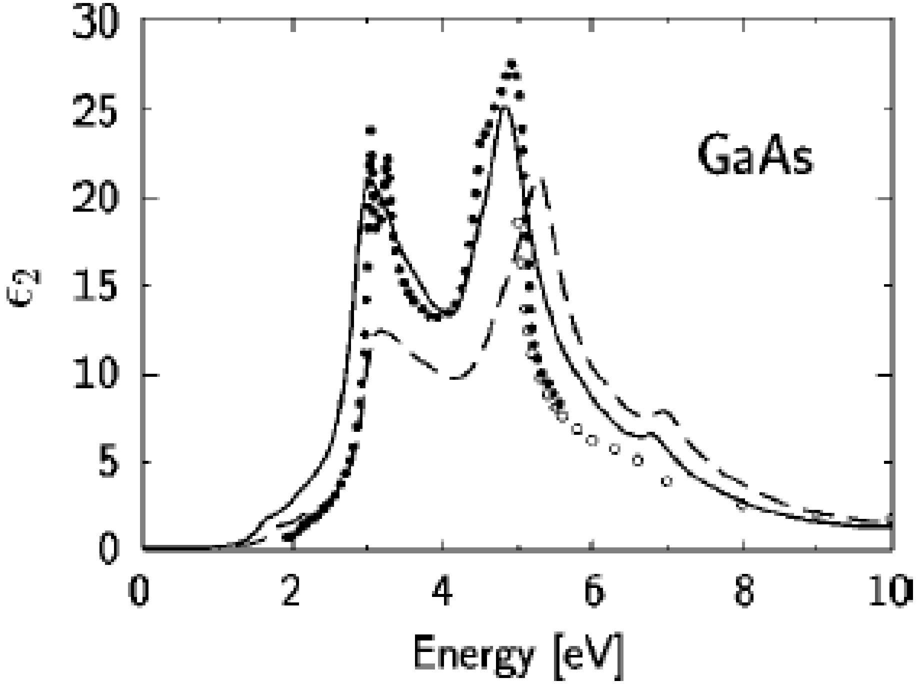
FIG. 2. Comparison of the calculated and experimental band structures for copper: solid line, *GW* quasiparticle excitation energies (Marini *et al.*, 2002); dashed line, DFT-LDA eigenvalues; ○, experimental data compiled by Courths and Hüfner (1984). A comparison to more recent experimental data (Strocov *et al.*, 1998, 2001) yields the same agreement.

Improve the single particle spectrum of electrons and holes.

1. Band structure
2. Band gap

2. Bethe-Salpeter equation : included the electron-hole interaction

Phys. Rev. B 62, 4927 (2000)



Improve the optical absorption spectrum.

FIG. 6. Calculated optical absorption spectrum of GaAs with (solid lines) and without (dashed lines) electron-hole interaction, using three valence bands, six conduction bands, 500 **k** points in the BZ, and an artificial broadening of 0.15 eV. The dots denote experimental data from Ref. 32 (○) and Ref. 33 (●).

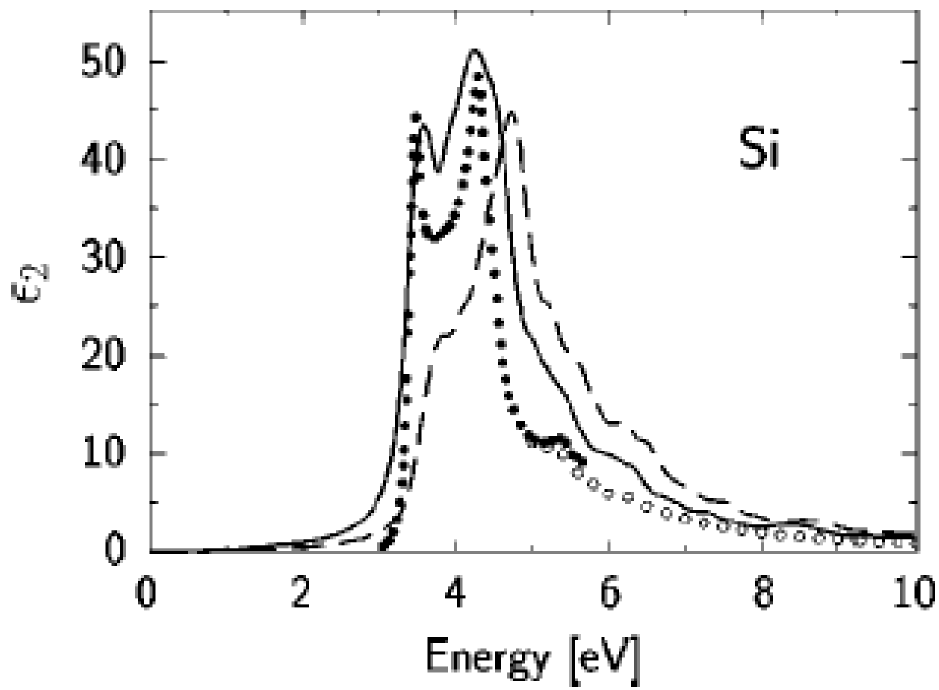


FIG. 8. Calculated optical absorption spectrum of Si with (solid lines) and without (dashed lines) electron-hole interaction, using three valence bands, six conduction bands, 500 **k** points in the BZ, and an artificial broadening of 0.15 eV. Experimental data are taken from Ref. 34 (○) and Ref. 35 (●).

Phys. Rev. B 62, 4927 (2000)

optics-vasp-modify

linear_optics.F main.F optics.F symmetry_orig.F

linear_optics_orig.F main_orig.F optics_orig.F symmetry.F

readme

readme

please replace main.F, optics.F and symmetry.F

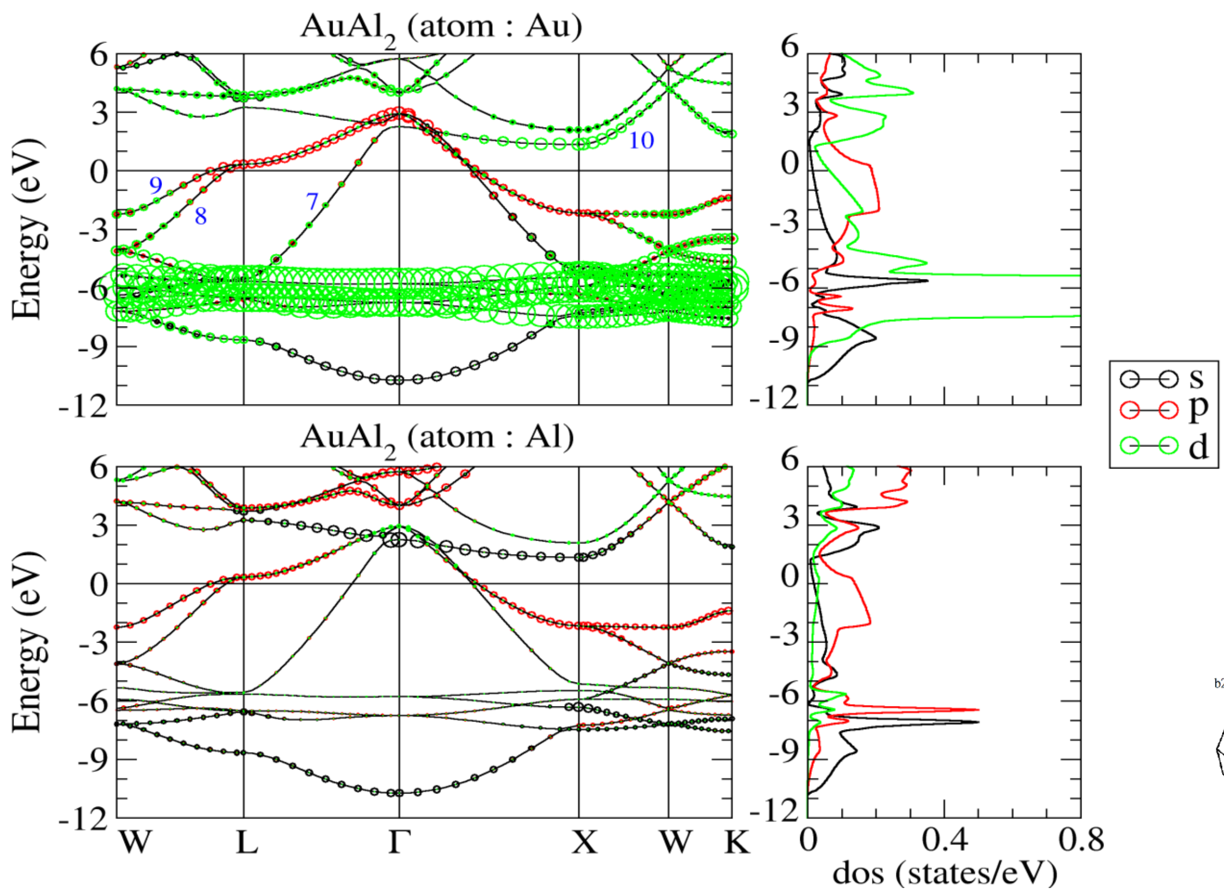
in the vasp.5.3.5 code with the corresponding subroutines

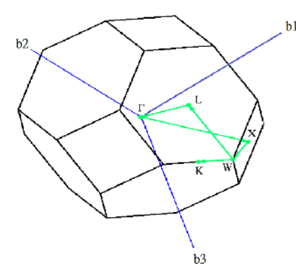
provided here. You can see the modifications I made

by comparing xxx.F and xxx_orig.F.

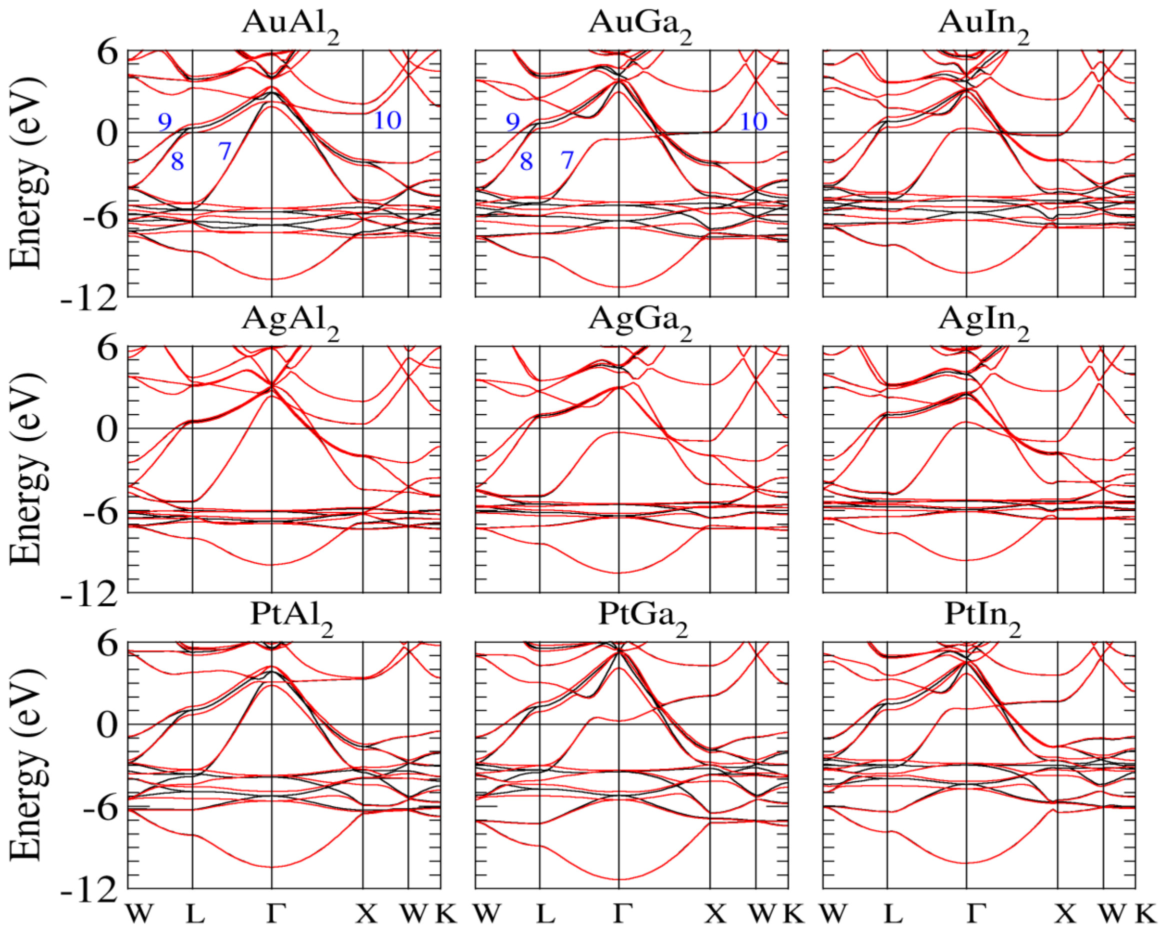
search the key word : "gyg" and "yao" to find the modifications

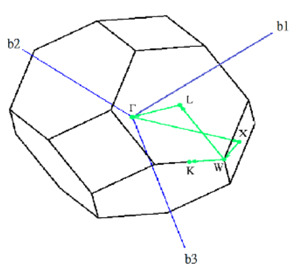
Band structure and density of states for $AuAl_2$



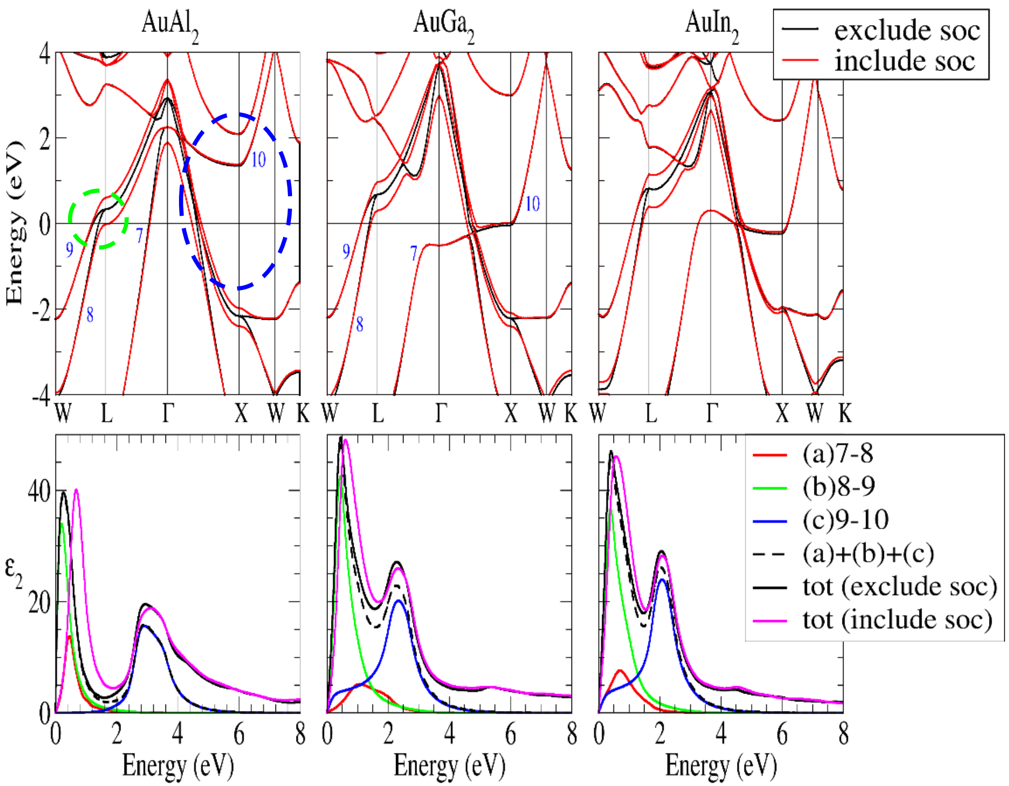


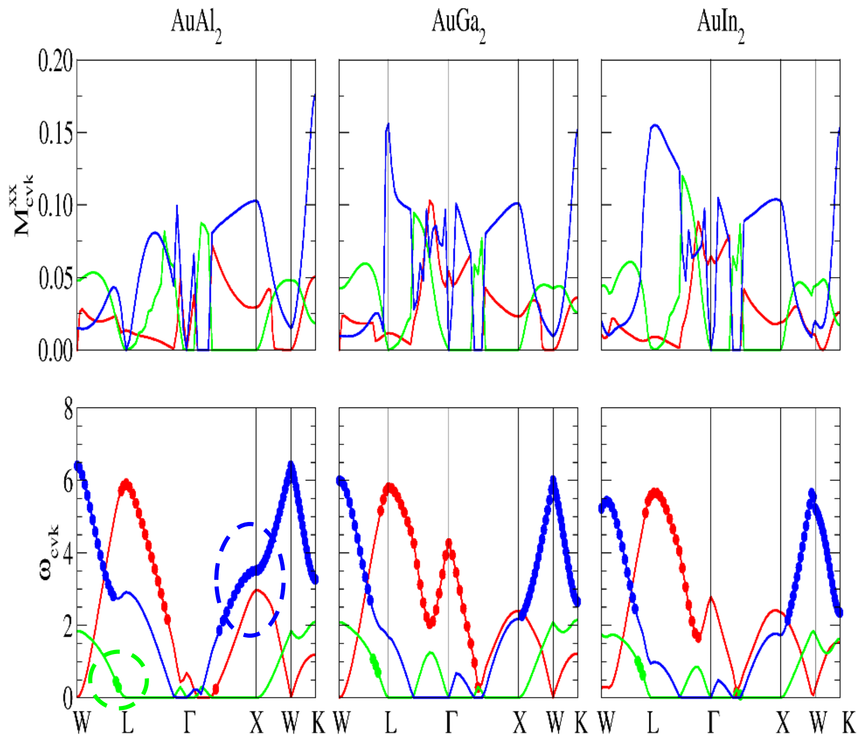
Band structure for noble-group III compounds





Interband transition ε_2 for Au-group III compounds





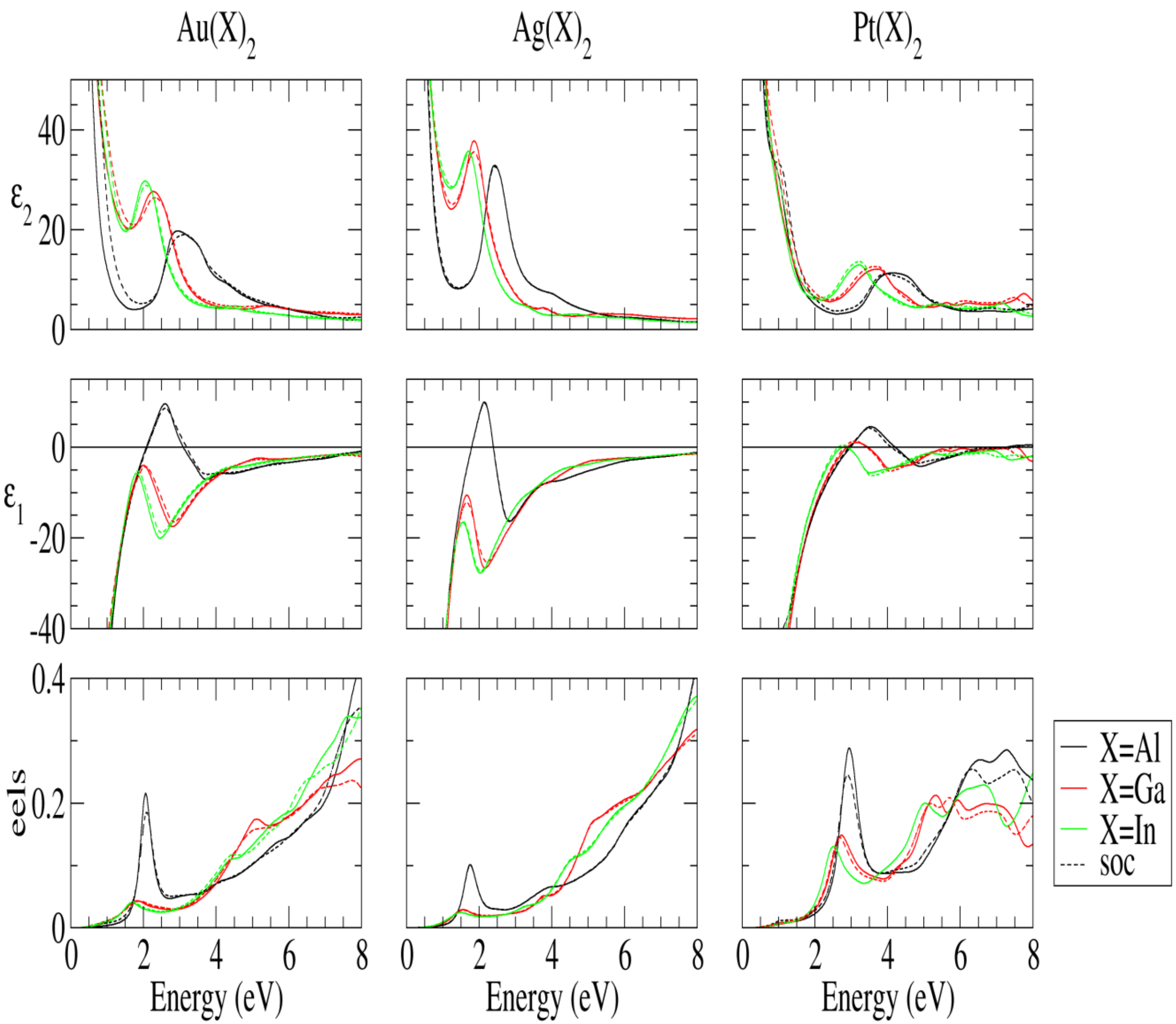
Intraband plasma energy Ω_p for noble-group III compounds

Ω_p (without SOC)					unit: eV
$PtAl_2$	8.09	$AuAl_2$	7.64	$AgAl_2$	7.86
$PtGa_2$	8.44	$AuGa_2$	7.72	$AgGa_2$	8.56
$PtIn_2$	8.01	$AuIn_2$	7.57	$AgIn_2$	8.22
Ω_p (with SOC)					
$PtAl_2$	7.64	$AuAl_2$	7.01	$AgAl_2$	7.76
$PtGa_2$	8.04	$AuGa_2$	7.29	$AgGa_2$	8.49
$PtIn_2$	7.69	$AuIn_2$	7.22	$AgIn_2$	8.12

$$\Omega^2_{p,\alpha\beta} \equiv \frac{4\pi e^2}{\Omega}\sum_{k}\sum_{n} 2w_k\delta(\varepsilon_{n,\mathbf{k}}-\varepsilon_f)(\hat{e}_\alpha \cdot \frac{\partial\varepsilon_{n,k}}{\partial\mathbf{k}})(\hat{e}_\beta \cdot \frac{\partial\varepsilon_{n,k}}{\partial\mathbf{k}})$$

$$\varepsilon_{\alpha\beta}(\omega) = 1 - \frac{\Omega^2_{p,\alpha\beta}}{\omega^2+i\Gamma\omega} \quad , \Gamma = 0.1 \text{ eV}$$

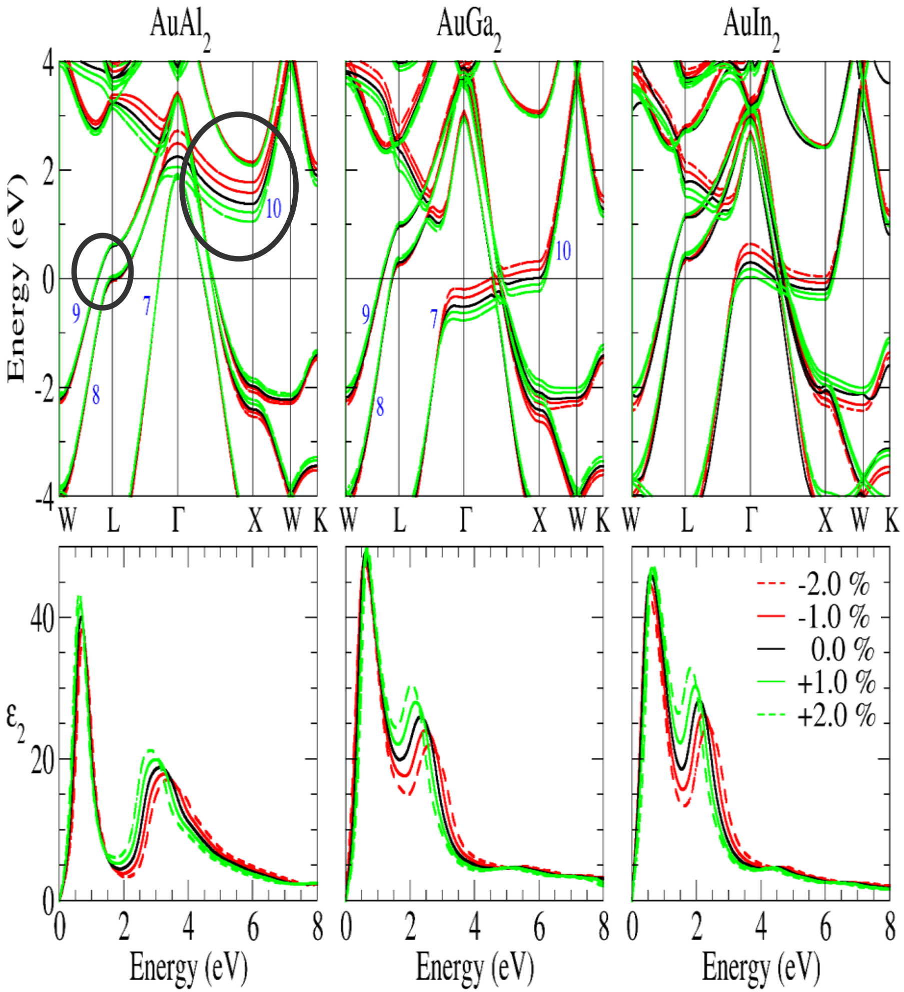
Dielectric function and EELS

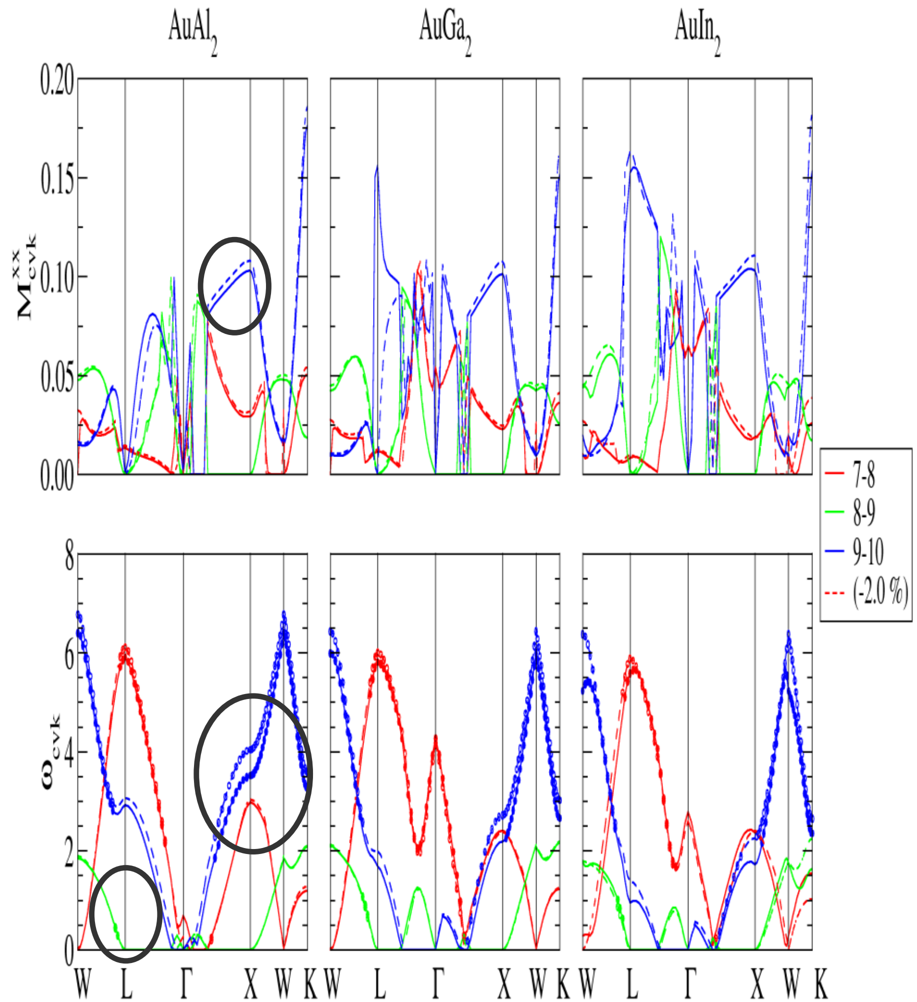


Bulk plasma energy ω_p

$AuAl_2$	2.08 eV
$AgAl_2$	1.76 eV
$PtAl_2$	2.90 eV
$PtGa_2$	2.68 eV
$PtIn_2$	2.46 eV

Strain effects on the interband contributions

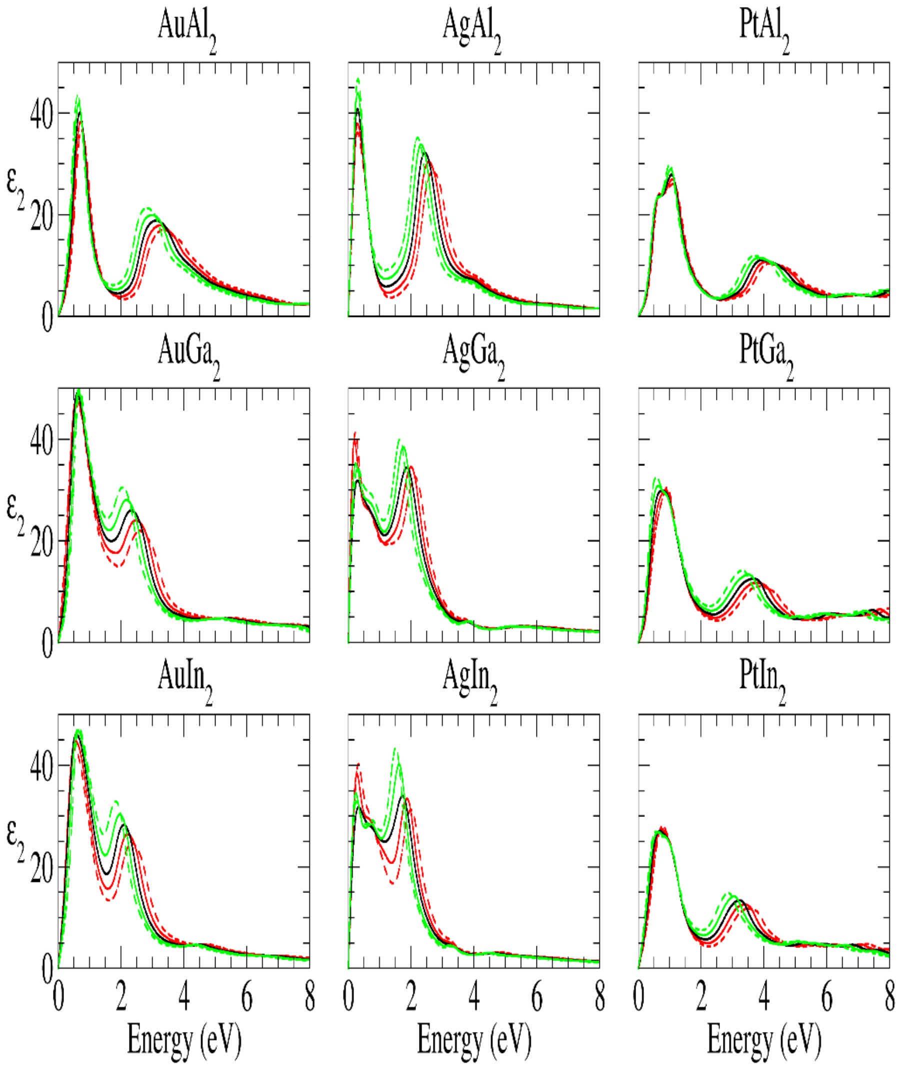


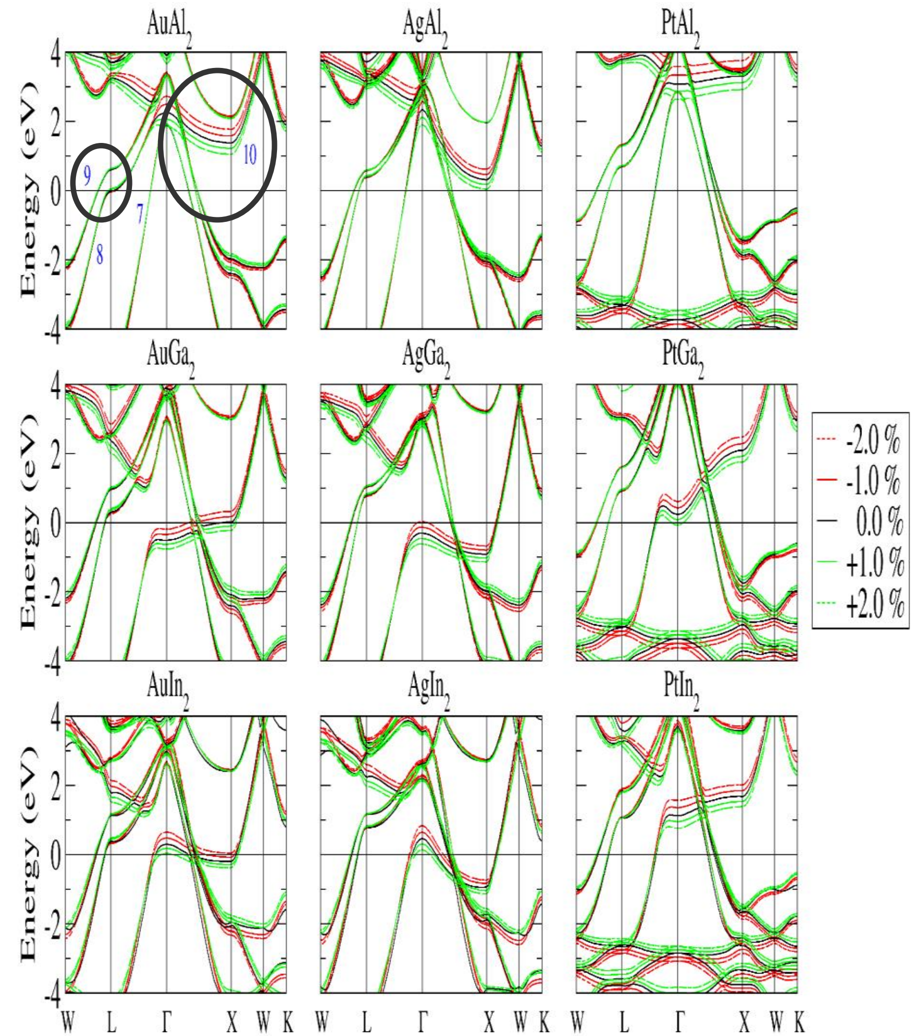


$\sigma = \sigma_x = \sigma_y = \sigma_z$

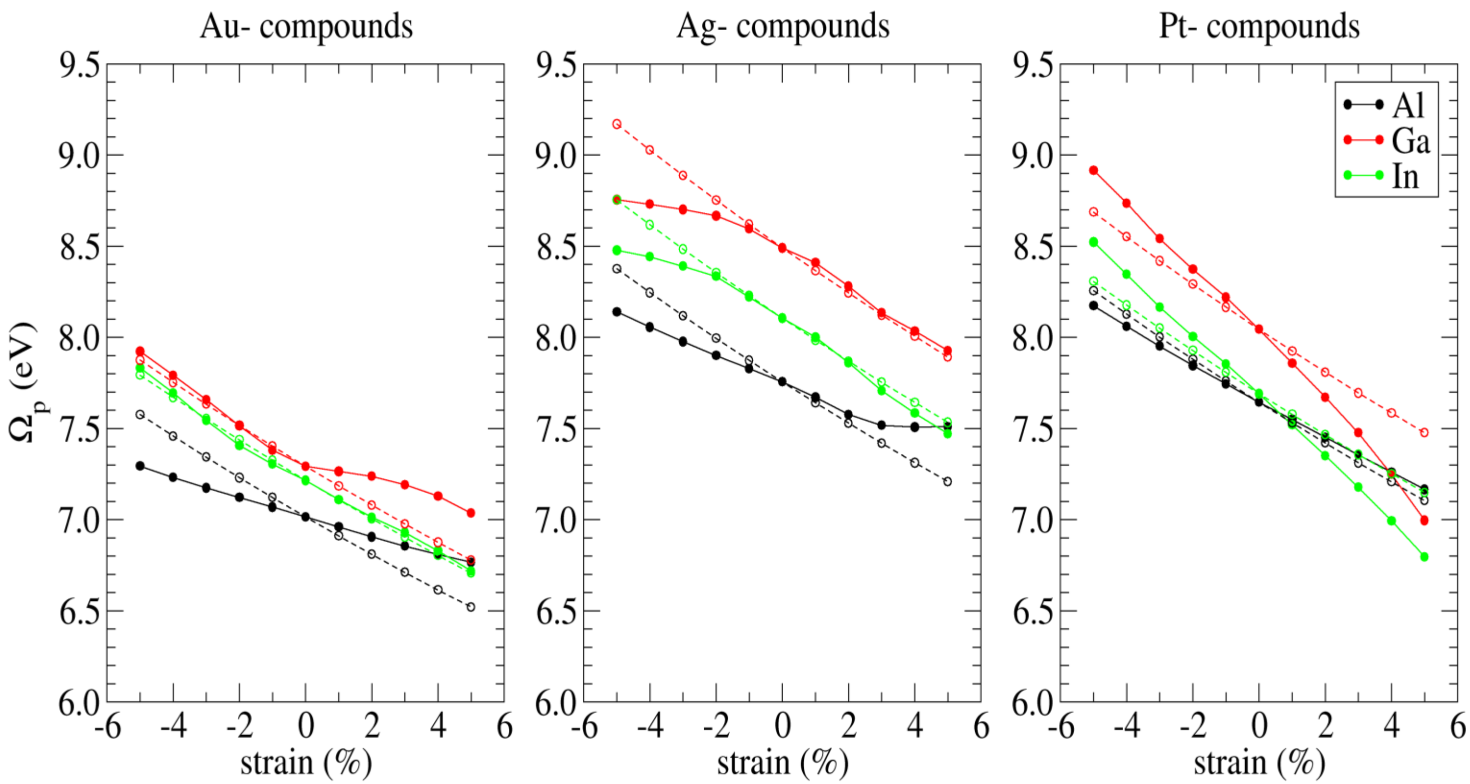
M^{xx}_{cvk} ⬆ ω^2_{cvk} ⬆

Strain effects on the interband contributions



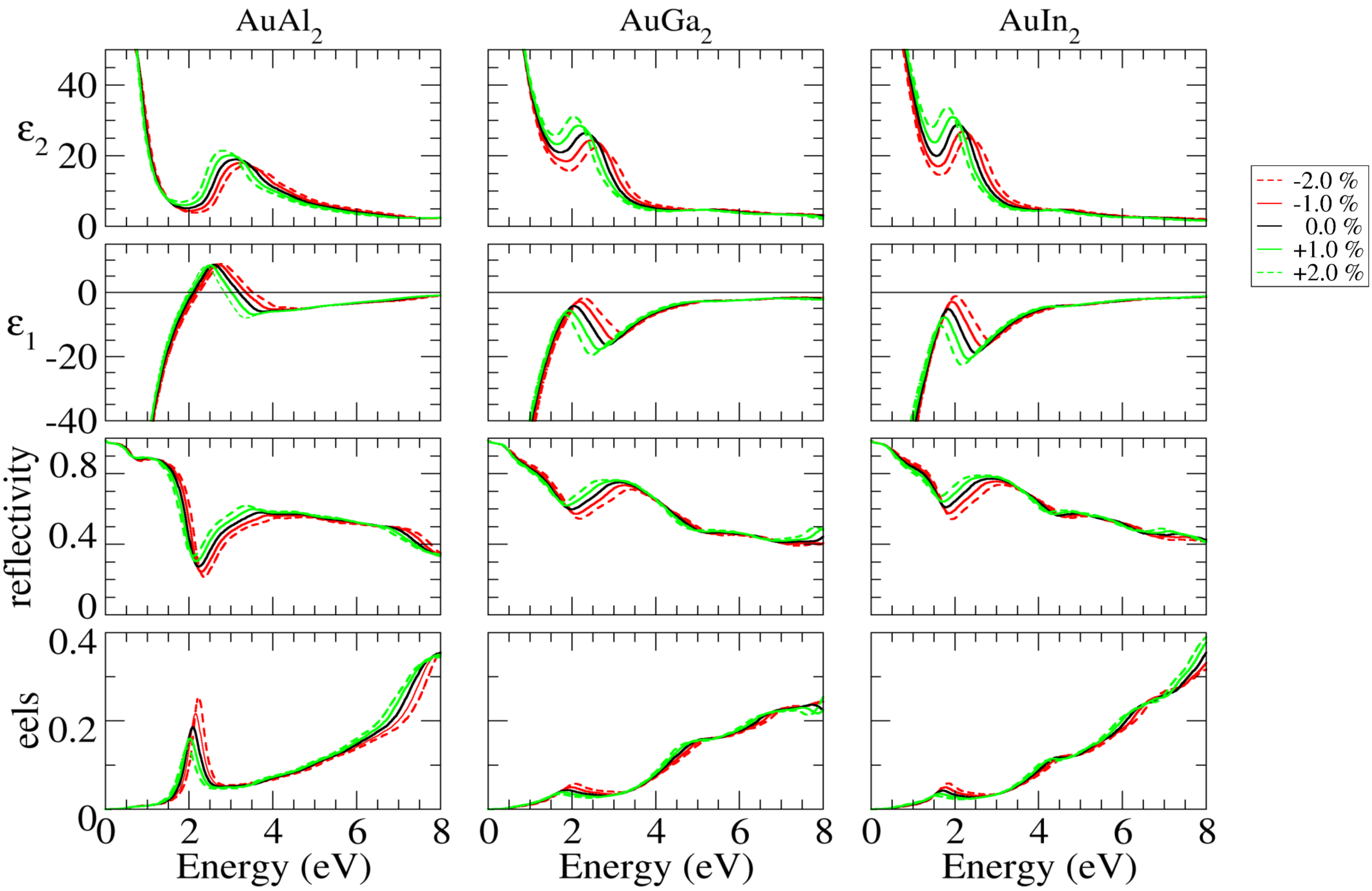


Strain effects on the intraband contributions



$$\Omega_p = \sqrt{\frac{4\pi n e^2}{m}} = \sqrt{\frac{4\pi N e^2}{mV}} \propto \sqrt{\frac{1}{V}}$$

Strain effects on the optical properties of Au-group III compounds



END