

Semiconductor dielectric function, excitons and the Penn model

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Abstract

Improved computation of the dielectric function considering excitonic effects and long wavelength is performed and compared with the nearly free electron band approximation, similarly with the Penn's model case. New expressions for the real and imaginary part of the dielectric function are presented and the real part compared with the Penn's result. The obtained functions satisfy the Kramers-Krönig relations, in contrast with earlier results in the literature. In addition, our improved dielectric function presents a coefficient of $2/3$ for small gap approximation (different from the value of 1 in the original Penn model) is very close to the value 0.62 obtained in [Can. J. Phys.53,(1975) p.2549] from pure numerical procedures. The obtained dielectric function also is used in a rough and stimative analysis of the metal-insulator transition in molecular hydrogen being the critical densities determinated near the experimental values for the hydrogen coming from other approach. The approximated expressions and critical values are given and the usefulness of the rough methods involved in the determination of the critical points briefly discussed.

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I. THE COMPLEX DIELECTRIC FUNCTION

As it is well known, the refractive index and the energy gap of semiconductors represent two fundamental aspects that characterize their optical and electronic properties. Penn in 1962 [1] proposed a simple model for an anisotropic semiconductor with electrons in a sphere of momentum space and by an isotropic energy gap. Penn's model, despite its simplicity, was and is the basic ingredient of several physical applications to diverse materials and different problems of great interests as the insulator-metal transition, exciton condensation and due to the isotropy of the system the model can be successfully applied to a liquid or an amorphous semiconductor. In this short report we improve the computation of the dielectric function going beyond the simple semiconductor considering the possibility of formation of excitons into the system. The obtained dielectric function (that fulfils the Kramers-Krönig relations [4]) is briefly analyzed and compared with the original Penn's one in its real (static) part in order to see the main differences and how the excitons modify the analytical properties of the dielectric function (critical points, singularities, zeroes, etc.) .

Our starting point is the expression for the complex dielectric function in the reduced zone [2]

$$\epsilon(\omega, q) = 1 - \lim_{\alpha \rightarrow 0} \frac{4\pi e^2}{q^2 \Omega} \sum_{k,l,l'} \frac{|\langle \bar{k}, l | e^{-iq \cdot r} | \bar{k} + \bar{q}, l' \rangle|^2 [f_0(E_{\bar{k}+\bar{q},l'}) - f_0(E_{\bar{k},l})]}{E_{\bar{k}+\bar{q},l'} - E_{\bar{k},l} - \hbar\omega + i\hbar\alpha}, \quad (1)$$

where $f_0(E_{\bar{k},l})$ is the distribution function for the reduced wave \bar{k} and the band l , Ω : volume

of the solid, $|\bar{k}, l\rangle$ is the Bloch wave function that satisfy $H_0 |\bar{k}, l\rangle = E_{\bar{k},l} |\bar{k}, l\rangle$ where H_0 is the Hamiltonian for an electron in the unperturbed lattice. $\epsilon(\omega, q)$ in the extended zone scheme takes the following form:

$$\epsilon(\omega, q) = 1 - \lim_{\alpha \rightarrow 0} \frac{4\pi e^2}{q^2 \Omega} \sum_{\bar{k}, \bar{G}} \frac{|\langle \bar{k} | e^{-iq \cdot r} | \bar{k} + \bar{q} + \bar{G} \rangle|^2 [N_{\bar{k} + \bar{q} + \bar{G}} - N_{\bar{k}}]}{E_{\bar{k} + \bar{q} + \bar{G}} - E_{\bar{k}} - \hbar\omega + i\hbar\alpha}, \quad (2)$$

where the distribution functions were changed by the respective zone occupation numbers.

The complex dielectric function is defined, however, as $\epsilon(\omega, q) = \epsilon_1(\omega, q) - i\epsilon_2(\omega, q)$

$$\epsilon_1(\omega, q) = 1 - \frac{4\pi e^2}{q^2 \Omega} \sum_{\bar{k}, \bar{G}} |\langle \bar{k} | e^{-iq \cdot r} | \bar{k} + \bar{q} + \bar{G} \rangle|^2 [N_{\bar{k} + \bar{q} + \bar{G}} - N_{\bar{k}}] \mathcal{P} \left(\frac{1}{E_{\bar{k} + \bar{q} + \bar{G}} - E_{\bar{k}} - \hbar\omega} \right), \quad (3)$$

$$\epsilon_2(\omega, q) = -\frac{4\pi e^2}{q^2 \Omega} \sum_{\bar{k}, \bar{G}} |\langle \bar{k} | e^{-iq \cdot r} | \bar{k} + \bar{q} + \bar{G} \rangle|^2 \delta(E_{\bar{k} + \bar{q} + \bar{G}} - E_{\bar{k}} - \hbar\omega). \quad (4)$$

We are interested in $\epsilon_1(0, q)$ and $\epsilon_2(\omega, q)$ in the Lim $q \rightarrow 0$ and to obtain closed expressions for these quantities from the above expressions (similarly as in the case of the Penn model for semiconductors).

The model is equivalent to the NFE (nearly free electron) model isotropically extended to 3 dimensions. The energies of the 2 bands are given by

$$E_{\bar{k}}^{\pm} = \frac{1}{2} \left\{ \left(E_{\bar{k}}^0 - E_{\bar{k}'}^0 \right) \pm \left[\left(E_{\bar{k}}^0 - E_{\bar{k}'}^0 \right)^2 + \overbrace{\left(E_g - E_x \right)^2}^{\Delta} \right]^{1/2} \right\} \quad (5)$$

where: $E_{\bar{k}}^0 = \frac{\hbar^2 k^2}{2m}$, $\bar{k}' = \bar{k} - 2k_F \hat{k}$, and $2k_F \hat{k}$ is the Fermi wave vector. Let us consider the wave functions $\psi_{\bar{k}}^{\pm}(\bar{r}) = \frac{e^{i\bar{k} \cdot \bar{r}}}{\Omega^{1/2}} \left[\frac{1 - \beta_{\bar{k}}^{\pm} e^{-i2k_F \hat{k} \cdot \bar{r}}}{(1 - \beta_{\bar{k}}^{\pm})^{1/2}} \right]$ where $\beta_{\bar{k}}^{\pm} = \frac{(E_g - E_x)}{2E_{\bar{k}}^{\pm}} = \frac{\Delta}{2E_{\bar{k}}^{\pm}}$, Ω is the volume and the signs $- (+)$ corresponds to the first (second) Brillouin zone. We suppose now (following the Srinivasan condition [3]) that \bar{k} and $\bar{k} + \bar{q}$ have the same reciprocal vector $-2k_F \hat{k} = \bar{G}$ (e.g. Umklapp process). Substituting into the general expressions (3,4), they depend now only on \bar{k}, \bar{q}

$$\epsilon_1(0) = 1 + \lim_{q \rightarrow 0} \frac{8\pi e^2}{q^2 \Omega} \sum_{\bar{k}} N_{\bar{k}} (1 - N_{\bar{k} + \bar{q}}) \frac{|\langle \bar{k} | e^{-iq \cdot r} | \bar{k} + \bar{q} \rangle|^2}{E_{\bar{k} + \bar{q}}^+ - E_{\bar{k}}^-} + \lim_{q \rightarrow 0} \frac{8\pi e^2}{q^2 \Omega} \sum_{\bar{k}} N_{\bar{k}} (1 - N_{\bar{k}' + \bar{q}}) \frac{|\langle \bar{k} | e^{-iq \cdot r} | \bar{k}' + \bar{q} \rangle|^2}{E_{\bar{k}' + \bar{q}}^+ - E_{\bar{k}}^-}, \quad (6)$$

and

$$\epsilon_2(\omega) = \lim_{q \rightarrow 0} \frac{4\pi e^2}{q^2 \Omega} \left[\sum_{\bar{k}} N_{\bar{k}} (1 - N_{\bar{k}+\bar{q}}) \left| \langle \bar{k} | e^{-iq \cdot r} | \bar{k} + \bar{q} \rangle \right|^2 \delta(E_{\bar{k}+\bar{q}}^+ - E_{\bar{k}}^- - \hbar\omega) + \right. \\ \left. + \frac{4\pi e^2}{q^2 \Omega} \sum_{\bar{k}} N_{\bar{k}} (1 - N_{\bar{k}'+\bar{q}}) \left| \langle \bar{k} | e^{-iq \cdot r} | \bar{k}' + \bar{q} \rangle \right|^2 \delta(E_{\bar{k}'+\bar{q}}^+ - E_{\bar{k}}^- - \hbar\omega) \right]. \quad (7)$$

Formally, we need to consider the following assumptions due to the specific physical problem that we are involved in:

i) ω is restricted to positive frequencies, then, $N_{\bar{k}} = 1$ iff $\bar{k} \in 1^{st}$ Brillouin zone, $N_{\bar{k}} = 0$ iff $\bar{k} \in elsewhere$,

ii) to perform the integrals, as usual, we assume a real crystal.

The evaluation of the matrix elements yields:

$$\left| \langle \bar{k} | e^{-iq \cdot r} | \bar{k} + \bar{q} \rangle \right|^2 = \frac{\left(\beta_{\bar{k}}^- + \beta_{\bar{k}+\bar{q}}^+ \right)^2}{\left[1 + (\beta_{\bar{k}}^-)^2 \right] \left[1 + (\beta_{\bar{k}+\bar{q}}^+)^2 \right]} \quad \text{and} \quad \left| \langle \bar{k} | e^{-iq \cdot r} | \bar{k}' + \bar{q} \rangle \right|^2 = \frac{\left(1 + \beta_{\bar{k}}^- \beta_{\bar{k}+\bar{q}}^+ \right)^2}{\left[1 + (\beta_{\bar{k}}^-)^2 \right] \left[1 + (\beta_{\bar{k}+\bar{q}}^+)^2 \right]}, \quad (8)$$

when $q \rightarrow 0$ the leading order of the matrix elements yields

$$\left| \langle \bar{k} | e^{-iq \cdot r} | \bar{k} + \bar{q} \rangle \right|^2 = \left| \langle \bar{k} | e^{-iq \cdot r} | \bar{k}' + \bar{q} \rangle \right|^2 \approx \\ \approx \frac{\cos^2 \theta (\Delta/4E_F)^2 (q/k_F)^2}{4 \left[(1 - k/k_F)^2 + (\Delta/4E_F)^2 \right]^2} \equiv \frac{\rho^2 x^2 \xi^2}{4(\eta^2 + \rho^2)^2}, \quad (9)$$

in the standard notation: θ is the angle between \bar{k} and \bar{q} , $E_F = \frac{\hbar^2 k_F^2}{2m}$, $\Delta = E_g - E_x$ and $\Delta/4E_F = \rho$. If the normal process does not contribute to $\epsilon_1(0)$ or $\epsilon_2(E)$, from previous expressions we have

$$\epsilon_1(0) = 1 + \frac{8\pi^2 e^2}{k_F^2 \Omega} \sum_{\bar{k}}^{1^{st} BZ} \frac{\rho^2 x^2}{4(\eta^2 + \rho^2)^2} \frac{1}{E_{\bar{k}+\bar{q}}^+ - E_{\bar{k}}^-}, \quad (10)$$

$$\epsilon_2(\omega) = \frac{4\pi^2 e^2}{k_F^2 \Omega} \sum_{\bar{k}}^{1^{st} BZ} \frac{\rho^2 x^2}{4(\eta^2 + \rho^2)^2} \delta(E_{\bar{k}}^+ - E_{\bar{k}}^- - \hbar\omega), \quad (11)$$

where the limit $q \rightarrow 0$ has been taken. The sum over \bar{k} (1^{st} Brillouin zone) is converted in an integral

$$\epsilon_1(0) = 1 + \frac{2E_p^2}{3\Delta^2} \left[(1 + \rho^2)^{1/2} - \rho \right], \quad E_p = \left(\frac{4\hbar^2 \pi n e^2}{m} \right)^{1/2}, \quad (12)$$

and

$$\epsilon_2(E) = +\frac{\pi E_p^2}{2E} \frac{[\Delta - \rho(E^2 - \Delta^2)^{1/2}]^2}{(E^2 - \Delta^2)^{1/2}}, \quad E = \hbar\omega, \quad (13)$$

the second equation holds for $\Delta \leq E \leq E_F(1 + \rho^2)^{1/2}$ and $\epsilon_2 = 0$ otherwise.

For Penn's case

$$\epsilon_1(0) = 1 + \frac{E_p^2}{E_g^2} \left[1 - (E_g/4E_F) + \frac{1}{3} (E_g/4E_F)^2 \right], \quad (14)$$

in our case

$$\epsilon_1(0) = 1 + \frac{2E_p^2}{3(E_g - E_x)^2} \left[\left(1 + \frac{(E_g - E_x)^2}{(4E_F)^2} \right)^{1/2} - \frac{E_g - E_x}{4E_F} \right], \quad (15)$$

clearly in this case there is a double pole (singularity) when $E_x \rightarrow E_g$ and we see the evident modification to the shape and critical points of the dielectric function due to the presence of excitons in the system. A full analysis considering jointly the quantum field theoretical part of the history will be present elsewhere [4]. For the imaginary part, when $\Delta^2 \rightarrow E^2$, $\epsilon_2(E) \rightarrow \infty$, and when $\Delta = 0$, $\epsilon_2(E) = 0$. In the next Section we will see the consistency of the above results making use of the Kramers-Krönig relations.

II. KRAMERS-KRÖNIG RELATIONS

Here we will corroborate the results of the previous paragraph by mean of the use of the Kramers-Kronig relations (following the same approximations as in previous Section). To begin with, from the relations between ϵ_1 and ϵ_2 we have

$$\epsilon_1(E) - 1 = \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\epsilon_2(E') E' dE'}{E'^2 - E^2}, \quad (16)$$

particularly for $E = 0$

$$\epsilon_1(E) - 1 = \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\epsilon_2(E') dE'}{E'}. \quad (17)$$

In the above equation the principal value symbol can be dropped: the delta function in $\epsilon_2(E')$ restricts its non-null range to $\Delta \leq E \leq E_F(1 + \rho^2)^{1/2}$. The crucial point now is the evaluation of the delta function in ϵ_2 up to the second order in q . From eq. (5) (previously to make the limit $q \rightarrow 0$) the energy difference is

$$E_{\vec{k}+\vec{q}}^+ - E_{\vec{k}}^- \approx E_F \left\{ 4(\eta^2 + \rho^2)^{1/2} - 2\eta \left[1 + (\eta^2 + \rho^2)^{-1/2} \right] \xi x + (\xi x)^2 (\eta^2 + \rho^2)^{-1/2} + \xi^2 \right\}, \quad (18)$$

with the definitions that were introduced before.

Now we convert the sum in \vec{k} in the first Brillouin zone in an integral obtaining

$$\epsilon_2(E) \approx \lim_{q \rightarrow 0} \frac{4k_F \rho^2 e^2}{2} \int_{-1}^1 \int_0^1 \frac{\rho^2 x^2}{(\eta^2 + \rho^2)^2} \delta(E_F W - E), \quad (19)$$

with

$$W \equiv \left\{ 4(\eta^2 + \rho^2)^{1/2} - 2\eta \left[1 + (\eta^2 + \rho^2)^{-1/2} \right] \xi x + (\xi x)^2 (\eta^2 + \rho^2)^{-1/2} + \xi^2 \right\},$$

where the lower limit in the η integration is zero. We must keep in mind here that q is small but not zero, then, we must retain the relevant order in q . We write the delta function as $\delta(f(x)) = \left| \frac{1}{df/dx} \right| \delta(x - x_0)$ with

$$f(\eta) = E_F \left\{ 4\eta (\eta^2 + \rho^2) - 2\eta \left[1 + (\eta^2 + \rho^2)^{-1/2} \right] \xi x + (\xi x)^2 (\eta^2 + \rho^2)^{-1/2} + \xi^2 \right\} - E \quad (20)$$

(where $E = \hbar\omega$)

As in several references based in the analytical models of semiconductors[6], we investigate the case $E = \Delta$

i) At $\eta = 0$, $f(\eta) > 0$ (because the limit $q \rightarrow 0$ has not been taken.)

ii) For $\eta \neq 0$, $4E_F(\eta^2 + \rho^2)^{1/2} - \Delta$ is positive and always dominates for sufficient small η (or q). Therefore, $f(\eta) > 0$ for $E = \Delta$ and the delta function cannot be satisfied.

This clearly means that the point $E = \Delta$ has no contribution to $\epsilon_2(E)$. Putting $f(\eta) = 0$ we determine η_0 in the limit $q \rightarrow 0$ in a similar manner that earlier works

$$\eta_0 = \eta_1 + \epsilon, \quad \epsilon \ll \eta_1 \quad (21)$$

$$\text{with } \eta_1 = \pm \left(\left(\frac{E}{4E_F} \right)^2 - \rho^2 \right)^{1/2}.$$

The expression for η_0 is valid for all $E > \Delta + O(\xi^n)$, $0 < n < 2$. The region of invalidity vanishes in the limit $\xi \rightarrow 0$ except in the precise point $E_g - E_x$. Finally we have

$$\delta(f(\eta)) \simeq \frac{(\eta^2 + \rho^2)^{3/2} \delta(\eta - \eta_0)}{E_F \left\{ 4\eta (\eta^2 + \rho^2) - 2 \left[\rho^2 + (\eta^2 + \rho^2)^{3/2} \right] \xi x + (\xi x)^2 \eta \right\}}, \quad (22)$$

putting all these ingredients in the expression for $\epsilon_2(E)$ we obtain

$$\epsilon_2(E) \approx \lim_{q \rightarrow 0} \frac{4k_F \rho^2 e^2}{2} \int_{-1}^1 \int_0^1 \frac{\rho^2 x^2}{(\eta^2 + \rho^2)^2} \frac{(\eta^2 + \rho^2)^{3/2} \delta(\eta - \eta_0)}{E_F \left\{ 4(\eta^2 + \rho^2)^{1/2} - 2 \left[\rho^2 + (\eta^2 + \rho^2)^{3/2} \right] \xi x + (\xi x)^2 \eta \right\}}, \quad (23)$$

we immediately obtain

$$\epsilon_2(E) = +\frac{\pi E_p^2}{2E} \frac{[\Delta - \rho(E^2 - \Delta^2)^{1/2}]^2}{(E^2 - \Delta^2)^{1/2}}, \quad E = \hbar\omega, \quad (24)$$

arriving to the same result as in the formula (13) of the previous Section. In an analogous manner and straightforwardly by mean eq. (16) and (24) we arrive to expression (12) as is required by consistency.

III. DIELECTRIC SCREENING AND INSULATOR METAL TRANSITION

As it is well known, one way to study the transition coming from the insulating phase, is supplied by the static dielectric function $\epsilon(0)$ of the molecular crystal which will be singular at the transition point because, as is expected, the transition to the metallic state is preceded with softning of some characteristic low lying excitation. At normal pressures and low temperature the "Frenkel exciton" is formed for a tightly bound electron hole pair.

In sharp contrast, when the pressure is considerably augmented to some critical value on the molecular crystal, the intermolecular interaction increases and the electron and the hole move further away from each other. Then, we can expect that at such high density in the non-conducting phase the excited state is a (much extended) Wannier exciton. In this case, the detailed lattice structure of the crystal is not very important to the pair and the common procedure is to simulate the lattice effects introducing effective electron and hole masses and screened Coulomb interaction $\sim 1/\epsilon r_{eh}$ to describe an isolated Wannier exciton. Then, the ground state of the energy of the electron hole pair is simply $-E_x$ (centre of mass kinetic energy term avoided) where

$$E_x = \frac{\mu}{m\epsilon^2}[R_y], \quad (25)$$

is the Wannier exciton binding energy. However, μ is the reduced mass, m is the bare electron mass and ϵ is the dielectric static function of the medium. In some earlier attempts, was claimed that is possible to use roughly this information to construct the corresponding effective model of the static dielectric function $\epsilon_1(0)$ of the medium on the low lying excitations in the case of compressed molecular hydrogen. We will follow such attempt in order to test it, due that our expression for the static dielectric function has a more detailed structure than the Penn's case (that is also the case of ref.[10]).

As we have computed, the static dielectric function (15) near the metal insulator transition $E_g \approx E_x$ becomes up to the lowest order

$$\begin{aligned} \epsilon_1(0) &\simeq 1 + \frac{2E_p^2}{3(E_g - E_x)^2} \left[1 + \frac{1}{2} \frac{(E_g - E_x)^2}{(4E_F)^2} - \frac{E_g - E_x}{4E_F} \right] \rightarrow \\ &\simeq 1 + \frac{2E_p^2}{3(E_g - E_x)^2} \dots\dots\dots, \end{aligned} \quad (26)$$

that clearly differs from the Penn case (and for the case of the ref.[10] for $E_g \gg E_x$ by a 2/3 factor into the second term. In practice this equation needs to be solved self-consistently: it is easy to solve the equation (26) if we know how μ and E_g simultaneously depend on $\epsilon_1(0)$ provided that the density limit of the existing electron-hole pair is maintained. Although the difficulty to find a full self-consistent solution for molecular hydrogen of $\epsilon_1(0)$ certainly exists, it is possible to obtain some estimates of the electric function near the insulator-metal transition. Consequently, our argument is that the exciton radius λ_x cannot be higher than a certain characteristic length $(q_{TF}^*)^{-1}$. Here q_{TF}^* is the critical Thomas-Fermi screening constant associated with a screened Heitler-London H_2 molecule immerse in an electron gas at fixed mean density prior dissociation: if the mean electron density of the gas is such that $q_{TF} > q_{TF}^*$ the screened H_2 molecule turns to be unstable. Using variational, perturbative and other methods [9] diverse values for q_{TF}^* (see Table I below) were found. These values can be used as upper limit for λ_x that is in the transition region

$$\lambda_x \simeq \lambda_x^* = \frac{m\epsilon}{\mu} q_{TF}^*[a.u], \quad (27)$$

If the neighborhood of the insulator-metal transition E_x varies slowly as function of the density in comparison with E_g we can substitute the result (27) back into (25) to obtain

$$E_x \simeq E_x^* = \frac{q_{TF}^*}{\epsilon} [Ry], \quad (28)$$

We see that near the insulator-metal transition E_x^* is saturated at this critical value. Using Friendly-Ashcroft band structure calculation [7] of the E_g for the molecular hydrogen under pressure together with the combination of (28) and (26), we get the following values of E_x^* for the diverse methods [9] used to obtain q_{TF}

<i>METHOD</i>		q_{TF}	$E_x^*[Ry]$	r_s^*
<i>Variational</i>	molecular orbital	1.197	0.067	1.56375
	electron pair	1.166	0.068	1.565
<i>Perturbative</i>		1.000	0.080	1.58
<i>Other(average)</i>		1.190	0.0672	1.564

Notice the lack of precision in r_s^* despite the different values of q_{TF} , $E_x^*[Ry]$ obtained by the different methods listed into the table above: evidently attempts as these [10] must be accompanied with other ones, looking for the microscopic/topological side of the exciton systems.

IV. CONCLUDING REMARKS

In this paper an improved derivation of the dielectric function based in a similar analytical model as the Penn proposal was presented, but now considering excitons in the system. Also in our calculation the Umklapp process as in [3] and long wavelength ($q \rightarrow 0$) was considered. The differences with Penn's model in the case of the real part of the dielectric function was given showing how the presence of excitons in the system modifies its behaviour. These results, being of a theoretical analytical character, were corroborated by mean of the Kramers-Krönig relations arriving independently to the same expressions for $\epsilon_2(E)$ and $\epsilon_1(E)$ up to the required order. Also it is important to note that our pure theoretical result presents a coefficient of 2/3 for small gap approximation (different from the value of 1 in the original Penn model) is very close to the value 0.62 numerically obtained by R. D. GRIMES and E. R. COWLEY in [11]. Using the improved dielectric function and making some standard assumptions about the behaviour of the exciton binding energies near a saturation value before the dissociation, we showed that $\epsilon_1(0)$ diverges at the transition point. This fact is in complete agreement with the experimental result of [8] and it stresses a discontinuous nature of the metal-insulator transition in solid hydrogen at low temperature under pressure. However same computation for different values of q_{TF} , $E_x^*[Ry]$ obtained by the different methods (listed in the Table) present a lack of precision showing the roughness of this type of approaches. However, the rough analysis of such a type of transitions must be complemented from a microscopic point of view. Consequently, it will be of particular interest to use in

near future the model presented here together with a "Keldysh-like" approach as [5] in order to analyze the metal insulator transition.

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