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INVESTIGATION OF THE ENERGY LOSS SPECTRUM OF THE FREE ELECTRON GAS: COMPARISON WITH COPPER

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The energy loss spectrum, ELS, with the aid of the Lindhard dielectric function for copper in the approximation of the free electron gas metal has been calculated and compared with that of the real copper. The zeros of the real part of the Lindhard dielectric function and that of the real metal are examined in the $(\boldsymbol{q}, \omega)$ plane. Also a table of results calculated for some parameters of "the free electron gas copper" is given.

I. INTRODUCTION

The Lindhard dielectric function [1] corresponds to the random-phase approximation (RPA). It provides a good description of the plasmon excitation modes and of long-wavelength screening phenomena.

Let us begin with the dielectric function of the electron gas. In the RPA it will be given by a scalar function of the magnitude of the wave number vector $\boldsymbol{q}$ [2]:

$$\mathscr{E}(\boldsymbol{q}, \omega) = 1 + \\ + \frac{e^2}{q^2 \Omega_e \epsilon_0} \sum_{\boldsymbol{k}} \frac{f(\boldsymbol{k}) - f(\boldsymbol{k}+\boldsymbol{q})}{E_f(\boldsymbol{k}+\boldsymbol{q}) - E_f(\boldsymbol{k}) - \hbar\omega + \mathrm{i}\alpha} \langle \boldsymbol{k} | \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}} | \boldsymbol{k}+\boldsymbol{q} \rangle \langle \boldsymbol{k}+\boldsymbol{q} | \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}} | \boldsymbol{k} \rangle \qquad (1)$$

in the limit $\alpha \to 0^+$, where Ω_e is the volume per electron, $f(\boldsymbol{k})$ is the occupation number of the state $\boldsymbol{k}$, $|\boldsymbol{k}\rangle = \frac{1}{\sqrt{\Omega_e}} \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}$ is the wave function corresponding to the energy $E_f(\boldsymbol{k}) = \frac{\hbar^2 k^2}{2m}$. Here, the matrix elements $\langle \boldsymbol{k} | \exp(-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}) | \boldsymbol{k}+\boldsymbol{q} \rangle$ and $\langle \boldsymbol{k}+\boldsymbol{q} | \exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}) | \boldsymbol{k} \rangle$ are equal to unity [2, 3]. Under these conditions, Lindhard obtained [1] the following analytical expression for the dielectric function of the electron gas in the RPA (in which only the diagonal elements are nonzero):

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$$\mathscr{E}_1(\beta, \delta) = 1 + \frac{K_s^2}{8\beta q^2}\left[\{1-(\beta+\gamma)^2\}\ln\left|\frac{1+\beta+\gamma}{1-\beta-\gamma}\right| + \{1-(\beta-\gamma)^2\}\ln\frac{1+\beta-\gamma}{1-\beta-\gamma} + 4\beta\right] \quad (2)$$

$$\mathscr{E}_2(\beta, \delta) = \frac{\pi K_s^2}{8\beta q^2}\begin{cases}\delta \text{ when } \beta < 1 \text{ and } \delta < |4\beta^2 - 4\beta| \\ [1-(\beta-\gamma)^2] \text{ when } |4\beta^2 - 4\beta| < \delta < |4\beta^2 + 4\beta|\end{cases}$$

$$0 \text{ when } \delta > |4\beta^2 + 4\beta| \quad (3)$$

$$= 0 \text{ when } \beta > 1 \text{ and } \delta < |4\beta^2 - 4\beta|,$$

where $\mathscr{E}_1$, $\mathscr{E}_2$ are the real and imaginary parts of the Lindhard dielectric function, respectively; $\beta = |q|/2k_F$, $\delta = \hbar\omega/E_F$, $\gamma = \delta/4\beta$; K_s is the inverse Fermi—Thomas screening length, k_F is the free-electron Fermi wave vector, and E_F is the Fermi energy.

The most important notions which are related with $\mathscr{E}$ are the density-fluctuation (excitation) spectrum (concerning the energy loss of a fast charge particle), and the time-dependent correlations between the density fluctuations (plasmon modes). The imaginary part of the inverse dielectric function,

$$-\operatorname{Im}\left(\frac{1}{\mathscr{E}}\right) = \mathscr{E}_2/(\mathscr{E}_1^2 + \mathscr{E}_2^2), \quad (4)$$

is proportional, for small $|\boldsymbol{q}|$, to the energy-loss function of a fast charged particle passing through the solid.

In the RPA, the dispersion of the plasma oscillations in the electron gas is given by the equation

$$\mathscr{E}(|\boldsymbol{q}|, \omega) = 0 \quad (5)$$

Therefore, the characteristic energy-loss spectrum has a sharp maximum at the frequency at which equation (5) is satisfied, and this frequency is known as the plasma frequency, ω_p.

The plasma frequency of the electron gas in the long-wave limit is given by the expression

$$\omega_p = (e^2 n/m\epsilon_0)^{1/2} \quad (6)$$

where n is the number of electrons per unit volume; ϵ_0 is the permittivity of vacuum (in SI units).

To date, there exist two basic experimental techniques for exciting and observing plasmons directly, and for obtaining their dispersion curve:

i) The passage of fast (keV) electrons through thin foils that produces longitudinal electric fields inside the sample and consequently excites plasmons.
ii) The optical detection of plasmons in thin films using p-polarized light.

In case of alkali metals, the situation is somewhat similar to the free-electron case. Haque and Kliewer [4] have found that the zero-wave-vector plasma frequency of sodium is smaller by about 4% and that of potassium smaller by about 19% than the corresponding free-electron values. Bross studied the energy-loss spectrum of aluminium [5] with the conclusion that Al is a nearly free-electron metal satisfying equation (5) for small $|\boldsymbol{q}|$'s.

It is the purpose of this paper to calculate the energyloss spectrum with the aid of the Lindhard dielectric function for copper as if it were a free-electron gas metal, and prove that the behaviour of real copper has little to do with the free-electron model, as its character is to be ascribed to the interband transitions from the 3d-bands to the empty bands [6]. A table is calculated for some parameters of the "free-electron gas Cu" in order to be used for related calculations.

Also the $|\boldsymbol{q}|$ and ω dependence of the zeros of $\mathscr{E}_1$ in the $(\boldsymbol{q}, \omega)$ plane is examined for both the "free-electron gas Cu" and the corresponding real metal. We have also studied the dependence of the ELS-peak height, H, and the "cut-off" energy on $|\boldsymbol{q}|$ in the case of the free-electron gas. The first has a sharp peak which corresponds to the first wave vector with $\mathscr{E}_1(|\boldsymbol{q}|, \omega) > 0$, namely, $\mathscr{E}_1(|\boldsymbol{q}| = 0.71\,\frac{2\pi}{a}, \omega)$. The "cut-off" energy, $\hbar\Omega$ (for any energy $E \geq \hbar\Omega$, the energy loss spectrum, $\operatorname{Im}(-1/\mathscr{E})$, vanishes) increases with $|\boldsymbol{q}|$.

II. RESULTS AND DISCUSSION

Both the real and the imaginary part, $\mathscr{E}_1$ and $\mathscr{E}_2$, of the Lindhard dielectric function are calculated according to equations (2) and (3), respectively, and then $\operatorname{Im}\left(-\frac{1}{\mathscr{E}}\right)$ is evaluated from equation (4). Table (1) gives the $|\boldsymbol{q}|$ — values in units of $2\pi/a$ for the calculated $\operatorname{Im}\left(-\frac{1}{\mathscr{E}}\right)$ in Figures 1—10. Our aim was to study the main differences between the energy -loss spectrum of the free-electron gas and that of copper. It is known from the analytical properties of the Lindhard dielectric function $\mathscr{E}(|\boldsymbol{q}|, \omega)$ and its asymptotic behaviour that:

$$\int_0^\infty \omega \operatorname{Im}\{\mathscr{E}(\boldsymbol{q}, \omega)\}^{-1}\, d\omega = -\frac{1}{2}\pi\omega_p^2 \quad (7)$$

Table 1

The absolute values of the wave vectors in units of $\frac{2\pi}{a}$ for which $-\,\mathrm{Im}\left(\frac{1}{\mathscr{E}}\right)$ are drawn in Figures 1—10.

q	$\lvert q \rvert$	q	$\lvert q \rvert$
⟨0.25, 0.25, 0⟩	0.3535		
		⟨0.75, 0, 0⟩	0.7500
⟨0.25, 0.25, 0.25⟩	0.4330	⟨0.5, 0.5, 0.5⟩	0.8660
		⟨1, 0, 0⟩	1.0000
⟨0.5, 0, 0⟩	0.5000	⟨1, 1, 0⟩	1.4142
⟨0.5, 0.5, 0⟩	0.7071	⟨1, 1, 1⟩	1.7321
		⟨2, 0, 0⟩	2.0000

Table 2

The "cut-off" energy $\hbar\Omega$ and the height of the ELS, H.

$q/(2\pi/a)$	$\hbar\Omega$(Ry.)	H
0.3535	0.58	0.12
0.4330	0.74	0.21
0.5000	0.87	0.36
0.7071	1.37	4.03
0.7500	1.47	2.31
0.8660	1.78	1.04
1.0000	2.18	0.61
1.4142	3.58	0.22

and that it satisfies the "perfect" screening requirement

$$\lim_{q \to 0} \langle \omega(\boldsymbol{q}, 0) \rangle^{-1} = 0 \qquad (8)$$

The main differences between the two ELS are:

a) If we insert the electron density of Cu, see Table (3), into equation (6), then $\hbar\omega_p = 0.79$ Ry. The centre of the predominant peak in the copper energy-loss spectrum is situated at 1.47 Ry [6], which indicates that copper is not a free-electron-like metal but the character of its ELS is due to interband transitions from the 3d-bands to the empty bands [6]; then condition (5) for the plasma oscillations is not satisfied. For this reason, sharp peaks are not expected in the copper ELS.

b) In the ELS of the free-electron gas, there are energies which we call the "cut-off" energies, $\hbar\Omega$, such that: for all energies $E \geq \hbar\Omega$ the $\mathrm{Im}(-1/\mathscr{E})$ vanishes. Table (2) gives the cut-off energies for several $|\boldsymbol{q}|$'s; this is drawn in Figure 12.

Table 3

Some parameters of the "free-electron gas copper" for $T = 0$ K

Electron concentration	$n = 4/a^3$ $= 8.25 \times 10^{22}$ cm
Fermi energy	$E_F = \frac{\hbar^2}{2m}(3\pi^2 n)^{2/3}$ $= 1.13 \times 10^{-18}$ Joule 7.06 eV.
Temperature of degeneracy	$T_E = \frac{2}{5}\frac{E_F}{k_B} = 32\,775$ K.
Radius of the Fermi sphere	$k(E_F) = (3\pi^2 n)^{1/3}$ $= 1.35 \times 10^8$ cm

Energy-level density at the Fermi energy per unit volume (spin degeneracy not taken into account)

$$D(E_F) = \frac{1}{4\pi^2}\left(\frac{2m}{\hbar^2}\right)^{3/2} E_F^{1/2} = \frac{m}{2\pi^2\hbar^2} k(E_F) = 5.59 \times 10^{39}\ \text{Joule}^{-1}\text{cm} = 8.96 \times 10^{20}\ \text{eV}^{-1}\text{cm}^{-3}$$

Specific heat of electrons per unit volume

$$C_V^{El} = \frac{2}{3}\pi^2 k_B^2 D(E_F)\, T = \gamma T = 70.2 \times 10^{-7}\ T\ \text{Joule/cm}^2\,\text{K}$$

Fermi velocity $v_F = \frac{\hbar k(E_F)}{m} = 1.56 \times 10^8$ cm/s.

Surface of the Fermi sphere $S = 4\pi k^2(E_F)$
$= 2.29 \times 10^{17}\ \text{cm}^{-2}$.

Maximum cross-section of the Fermi sphere

$$A_{max} = S/4 = 0.57 \times 10^{17}\ \text{cm}^{-2}.$$

Cyclotron frequency of electrons in a magnetic field B

$$\omega_c = \frac{e}{m} B = 2.21 \times 10^7 \left(B \left[\frac{\text{A}}{\text{cm}} \right] \right) \text{s}^{-1}$$

Classical cyclotron radius for electrons at the Fermi energy

$$R_c = \frac{v_F}{\omega_c} = \frac{7.05}{B[\text{A/cm}]}\ \text{cm}.$$

Electrical conductivity of electrons having the Fermi energy E_F and a mean free patrh Λ:

$$\sigma = \frac{ne^2}{mv_F}\Lambda = 1.38 \times 10^{11}\ (\Lambda\ [\text{cm}])\ \Omega^{-1}\text{cm}^{-1}$$

Fermi—Thomas screening wave vector

$$K_s = (6\pi\, ne^2/E_F)^{1/2} = 1.78 \times 10^8\ \text{cm}^{-1}.$$

The vanishing of the ELS for copper is not seen till 10 Rydberg [7, 8]. From Table (2), one can see that the height of the Lindhard ELS increases till $|\boldsymbol{q}| = 0.71\left(\frac{2\pi}{a}\right)$, which is the first wave vector with $\mathscr{E}_1(|\boldsymbol{q}|, \omega) > 0$ (i.e. without zeros in the real part of the Lindhard dielectric function). In the case of copper the maximum height of the main ELS peak is about 1 as $\boldsymbol{q} \to 0$ and decreases with increasing $|\boldsymbol{q}|$ [3, 6].

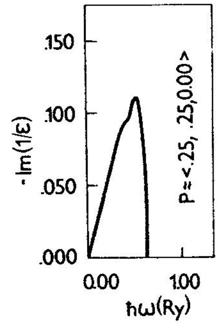


Fig. 1. The energy-loss spectrum, $-\mathrm{Im}(\mathscr{E}^{-1}(\boldsymbol{q}, \omega))$, for copper as if it were a free-electron gas metal for ten wave vectors ($\boldsymbol{p} = \boldsymbol{q}$) in the units of $\frac{2\pi}{a}$ for energies $0 \le \hbar\omega \le 4$ Ry. $|\boldsymbol{q}| = 0.3535$.

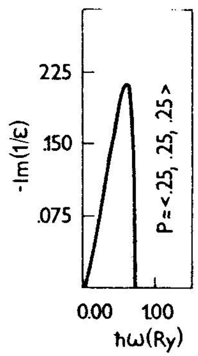


Fig. 2. as Fig. 1 for $|\boldsymbol{q}| = 0.4330$

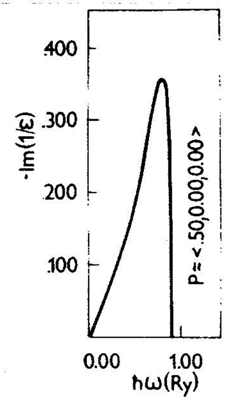


Fig. 3. as Fig. 1 for $|\boldsymbol{q}| = 0.5000$

c) For $|\boldsymbol{q}| \ge \sqrt{3}\left(\frac{2\pi}{a}\right)$, there are intervals $[0, \omega_{th}(|\boldsymbol{q}|)]$, at which $\mathscr{E}_2(|\boldsymbol{q}|, \omega_{th}(|\boldsymbol{q}|))$, or $\mathrm{Im}(-1/\mathscr{E})$, vanishes, where ω_{th} is a "threshold" frequency. Such intervals are

only found in the case of copper when $\boldsymbol{q} = \boldsymbol{K}$[3, 10], where $\boldsymbol{K}$ is a reciprocal lattice vector, as a result vanishing of the intraband contributions in the calculations of $\mathscr{E}$ $(\boldsymbol{K}, \omega)$.

d) When $\boldsymbol{q}$ is large enough (i.e. comparable with reciprocal lattice vectors), then $\mathrm{Im}(-1/\mathscr{E}) \approx \mathscr{E}_2$, because in such cases $\mathscr{E}_1 \approx 1$ and $\mathscr{E}_2$ is small. Such straightforward behaviour of the ELS is also known in the case of copper when the core electrons are excited.

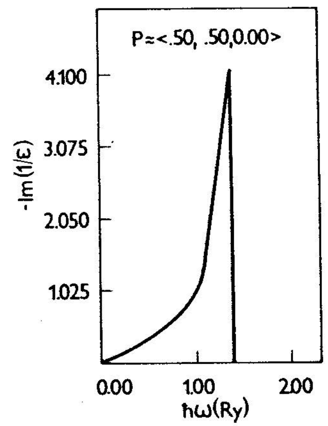


Fig. 4. as Fig. 1 for $|\boldsymbol{q}| = 0.7071$

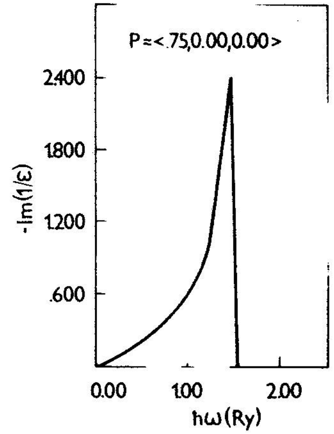


Fig. 5. as Fig. 1 for $|\boldsymbol{q}| = 0.7500$

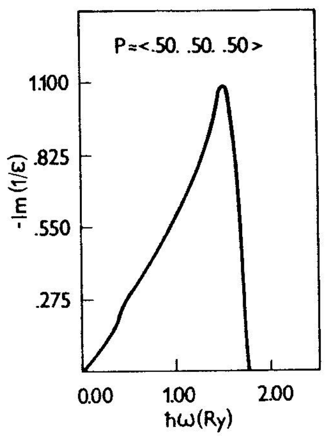


Fig. 6. as Fig. 1 for $|\boldsymbol{q}| = 0.8660$

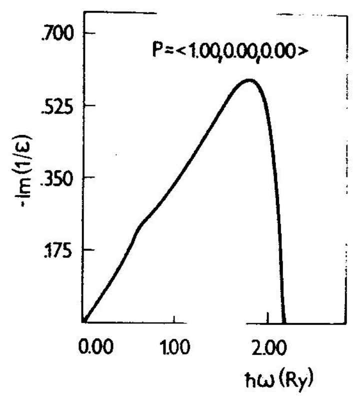


Fig. 7. as Fig. 1 for $|\boldsymbol{q}| = 1.0000$

Fig. 11 shows the dependence of both zeros, ω_0 and $\tilde{\omega}$, of $\mathscr{E}_1(\boldsymbol{q}, \omega)$ on $|\boldsymbol{q}|$ for Cu [3] and the free-electron gas.

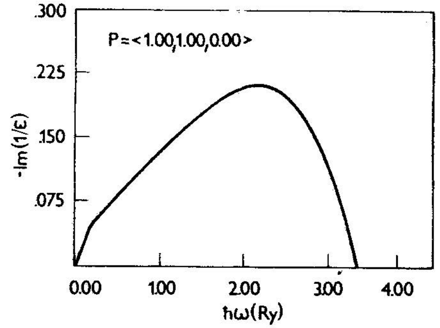


Fig. 8. as Fig. 1 for $|\boldsymbol{q}| = 1.4142$

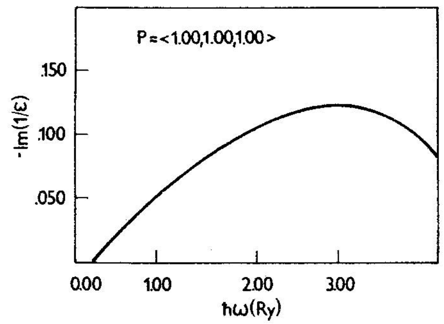


Fig. 9. as Fig. 1 for $|\boldsymbol{q}| = 1.7321$

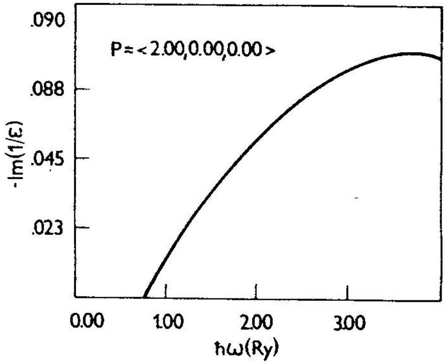


Fig. 10. as Fig. 1 for $|\boldsymbol{q}| = 2.0000$

For a small $|\boldsymbol{q}|$, the difference in the Fermi factors in equation (1) becomes [1]:

$$f(\boldsymbol{k}+\boldsymbol{q}) - f(\boldsymbol{k}) \approx \boldsymbol{q}.\frac{\partial f}{\partial \boldsymbol{k}} \approx q\mu\delta(|\boldsymbol{k}| - k_f), \tag{9}$$

$$\mu = \boldsymbol{k}.\boldsymbol{q}/|\boldsymbol{k}|\,|\boldsymbol{q}|. \tag{10}$$

Dropping the terms of the order of magnitude q^2 in the intergrand, the dielectric function becomes

$$\varepsilon(|\boldsymbol{q}|, \omega) = 1 + \frac{K_s^2}{2|\dot{\boldsymbol{q}}|^2}\int_{-1}^{1}\frac{\mu\,d\mu}{\mu-\gamma}$$
$$= 1 + \frac{K_s^2}{2|\dot{\boldsymbol{q}}|^2}\left(2 - \gamma\ln\left|\frac{1+\gamma}{1-\gamma}\right|\right), \tag{11}$$

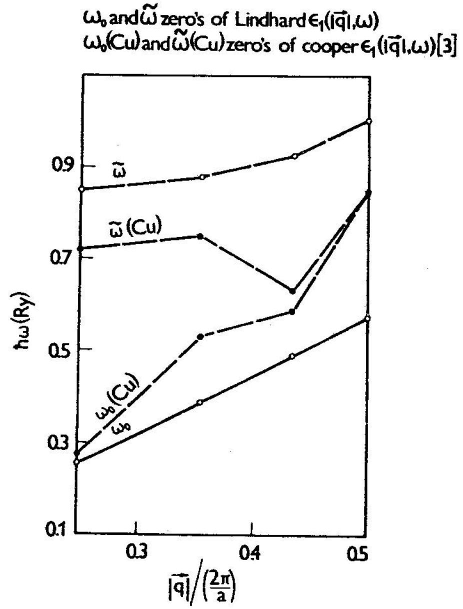
where $\gamma = \omega/|\boldsymbol{q}|\,v_F$. The condition of the lower zero, ω_0, which looks like a damped transverse-like mode in the continuum, arises when

$$2 \approx \gamma\ln\left|\frac{1+\gamma}{1-\gamma}\right|. \tag{12}$$

This condition requires a fairly linear $\omega_0(|\boldsymbol{q}|)$ curve (see Fig. 11), i.e.

$$\omega_0 \approx \frac{5}{6}v_F|\boldsymbol{q}|. \tag{13}$$

From Fig. 11, the two curves of the free-electron gas tend to merge at $|\boldsymbol{q}| = 0.71\left(\frac{2\pi}{a}\right)$, but those of Cu merge at $|\boldsymbol{q}| \approx 0.5\left(\frac{2\pi}{a}\right)$. It can be seen from Figure 11 that the copper lower zero does not result from a linear dispersion curve. This is expected for the 3*d*-metals.



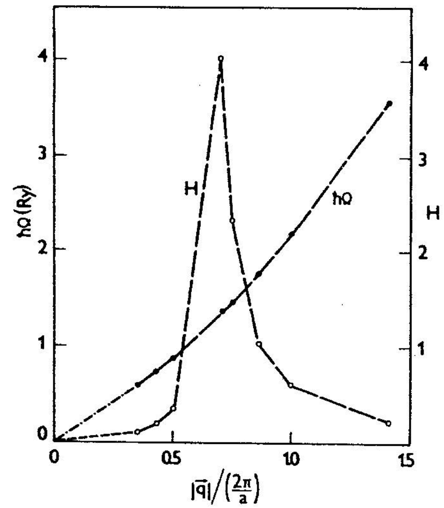


Fig. 11. Position of the zeros of $\mathcal{E}_1(\boldsymbol{q}, \omega)$ for copper and a free-electron gas in the $(\boldsymbol{q}, \omega)$ plane, where ω_0, $\tilde{\omega}$ are zeros of the Lindhard function $\mathcal{E}_1(|\boldsymbol{q}|, \omega)$ and $\omega_0(\mathrm{Cu})$, $\tilde{\omega}(\mathrm{Cu})$ the corresponding zeros of copper.

Fig. 12. The variation of the "cut-off" energy, $\hbar\Omega$, and the height of $-\mathrm{Im}(1/\Sigma)$, H, with $|\boldsymbol{q}|/\left(\frac{2\pi}{a}\right)$.

Pippard [9] has given data of an idealized metal on the basis of the free-electron model. In his model the electron concentration is assumed to be $n = 6 \times 10^{22}\ \mathrm{cm}^{-3}$. We give here (in Table 3) some parameters for copper, as if it were a free-electron gas metal, for its use in the empty-lattice test and for comparison with the data of real copper. The copper lattice constant, a, has the value 6.816 a.u(1 a.u = 0.0529).

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УЗУЧЕНИЕ СПЕКТРА ЭНЕРГЕТИЧЕСКИХ ПОТЕРЬ СВОБОДНОГО ЭЛЕКТРОННОГО ГАЗА: СЛУЧАЙ МЕДИ

В работе приводятся результаты расчета спектра энергетических потерь, полученного при помощи диэлектрической функции Линдгарда для меди в приближении свободного электронного газа в металле, а также их сравнение с реальными данными для меди. В плоскости (q, ω) изучены нули диэлектрической функции Линдгарда, а также нули, полученные в результате измерений для меди. Кроме того, приводится таблица некоторых параметров, расчитанных в приближении свободного электроного газа в меди.