Ultraviolet and soft X–ray photon–photon elastic scattering in an electron gas.

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We have considered the processes which lead to elastic scattering between two far ultraviolet or X–ray photons while they propagate inside a solid, modeled as a simple electron gas. The new ingredient, with respect to the standard theory of photon–photon scattering in vacuum, is the presence of low–energy, nonrelativistic electron–hole excitations. Owing to the existence of two–photon vertices, the scattering processes in the metal are predominantly of second order, as opposed to fourth order for the vacuum case. The main processes in second order are dominated by exchange of virtual plasmons between the two photons. For two photons of similar energy $\hbar\Omega$, this gives rise to a cross section rising like $\Omega^2$ up to maximum of around $10^{-32}$ cm$^2$, and then decreasing like $\Omega^{-6}$. The maximal cross section is found for the photon wavevector $k \sim k_F$, the Fermi surface size, which typically means a photon energy $\hbar\Omega$ in the keV range. Possible experiments aimed at checking the existence of these rare but seemingly measurable elastic photon–photon scattering processes are discussed, using in particular intense synchrotron sources.
I. INTRODUCTION

Photon–photon scattering processes are well known in vacuum. The lowest order perturbation theory diagrams are shown on Fig.1. The corresponding scattering amplitude and cross section, first calculated by Karplus and Neumann [1], were later thoroughly investigated by de Tollis [2], and are extensively reviewed in Lifshitz and Pitaevskii’s book [3]. The cross section of the process in vacuum turns out to be very small, of the order of magnitude of $10^{-30} \text{cm}^2$ at the threshold for the production of real $e^- - e^+$ pairs in the intermediate state, which corresponds to an incident photon energy of $\hbar \Omega = mc^2 \sim 0.5 \text{ MeV}$. For energies $\hbar \Omega \ll mc^2$, the cross section falls as $\sigma \sim (\frac{\Omega}{mc^2})^6$. It is thus extremely small in the eV range, where powerful laser beams are available, or even in the keV range, where strong synchrotron sources are now feasible. The physical reason is clear: if the energy of photons lies too far below the ”energy gap” $2mc^2$, the virtual intermediate $e^- - e^+$ pairs are hard to excite, and this greatly cuts the probability of the process. In the end, this makes the process very difficult to pursue experimentally – albeit not impossible [11], [12].

One is thus led to the obvious idea that, if instead of vacuum, the photons were embedded in a material medium, where pair excitations could be created with a smaller or zero energy gap, then the probability of photon-photon scattering in such a system might be enhanced.

Condensed matter physics provides us with countless examples of systems with this property: electron–hole pairs replace electron–positron pairs, and this reduces excitation energies from MeV to the eV or meV range. For example, in a semiconductor the intermediate state involves pairs excited across the semiconducting band gap.

Third order nonlinear optical susceptibilities, which in principle involve the same diagrams of Fig.1, have for example been well characterized, both experimentally [4], and theoretically [5], [6], for the quasi–one–dimensional prototypical semiconductor polyacetylene.

The focus of this paper will be however on the elastic scattering cross section among very high energy photons, which makes the problem radically different from those studied so far.
To make a start, we would like to study photon–photon scattering inside the simplest and most typical condensed system, for example that of a simple metal. Of course, even simple metals have in real life a number of complications. The periodic potential brings about a multi–band problem. Due to the multiplicity of bands, interband transitions may occur, etc. However we shall restrict in this paper to the contribution of intraband transitions, right at the Fermi surface, and these processes can be modelled with a single parabolic band. Moreover, in the keV photon energy range (which will turn out to be the most interesting case) virtually all systems, including insulators, can often be treated as free electron metals to a very good approximation.

A second complication is that in a metal, or more generally in a solid or liquid, photons are heavily absorbed. Their energy decays into electron–hole pairs, and finally into heat. Of course, this fact may represent a practical obstacle at getting strong photon fields inside an actual material. Yet, for example, one can think of using a very intense evanescent photon field at a surface in a reflection experiment, or use intense, far ultraviolet and X–ray beams from a synchrotron radiation source, tuned so that the the absorption coefficient is small enough to be neglected.

A third relevant issue might be the difficulty of separating elastic from inelastic photon–photon processes, when the process takes place inside a metal. Since there is no gap, a shower of soft electron–hole pairs is likely to accompany the photon–photon collision, and it could be difficult in practice to separate this inelastic part from the elastic process we are after.

Postponing further discussion of some of these, and other related issues to a later section of this paper, we will now proceed with the actual calculation of the elastic photon–photon cross section in an electron gas, so as to come up with predictions for orders of magnitude and angular dependences. This calculation is not simply a generalization of the classic calculation of \[ \| \] from vacuum to a uniform electron gas. The nonrelativistic nature of the electron gas allows for new processes, which do not exist in vacuum. There are new energy scales, the Fermi energy \( E_F \) and the plasma energy \( \hbar \omega_p \), which are absent in vacuum.
Anticipating our later results, we will end up with numerical values of the cross section, which are again exceedingly small - we estimate for a potassium target a peak value of $10^{-32}$ cm$^2$, somewhat smaller than that in vacuum. However, the peak is now in the keV photon frequency range. Since ultraviolet and soft X–ray photon sources are so much more intense than γ–ray sources, the possibility that our calculations might not remain academic seems real. Moreover, our estimated photon–photon cross section in the ultraviolet is $\sim 10^{-35}$ cm$^2$, which might be measurable using medium power lasers. At this stage, it is not clear to us whether such measurements are going to be feasible, or worth doing, or whether the present calculation will remain an academic exercise. We simply would like to point out that the new photon-photon processes exist, and that they are in principle finite and measurable.

As announced, we shall take the uniform, single–band non–relativistic electron gas (EG) as our model for the metal. The model is completely specified by the electron mass $m$ and the Fermi energy $E_F$. In section 2 we identify the simplest possible photon–photon scattering processes, which now appear in the second, third and fourth order of the perturbation theory (as opposed to just fourth order in vacuum). In later sections, we choose two of these processes and calculate them out in detail. First, in section 3 we deal with the 2nd order processes, which are the largest, and consist essentially of exchange of plasmons between the photons. These processes do not exist in vacuum. In section 4 we then investigate the 4th order process, formally the same square diagram of Fig.[1]. Now, however, positrons are replaced by holes in the Fermi sea, which modifies both the form and the magnitude of the resulting scattering amplitude. The resulting fourth–order cross section, not surprisingly, turns out in the end to be many orders of magnitude smaller than that of the 2nd order processes, even at very high frequencies (where they could in principle have come closer). We therefore identify plasmon exchange as the dominant mechanism for the photon-photon scattering in the EG. In section 5 we summarize our results, and discuss briefly the possibilities of experimentally observing the proposed photon–photon elastic scattering processes.
II. POSSIBLE SCATTERING PROCESSES

Before moving on to the actual calculation, we wish to stress some points, which make the situation of photons inside the EG different from that in vacuum. First of all, a photon in an electron gas is a well-propagating mode only at frequencies much larger than the plasma frequency $\omega_p$. Approaching that frequency from above, photons become dressed, absorption increases, until below $\omega_p$ photons do not propagate anymore, and get absorbed within a wavelength or so. In an actual experimental situation, a beam from an external source penetrates into the metal, and the scattered, reflected or transmitted radiation, which leaves the metal, is detected. In these conditions, what actually scatters inside the metal, are the dressed photons, as they enter through the surface. To predict the outcome of such experiment rigorously, one would have to perform the calculation including photon dressing, plus the effect of the surfaces. Both represent nontrivial complications, which we shall not endeavor to consider at this stage. Therefore, we simply assume here free photons propagating in an infinite bulk electron gas. We shall restrict our treatment to frequencies $\Omega$ higher than $\omega_p$

$$\hbar \Omega \sim \hbar \omega_p,$$  \hfill (1)

and completely neglect the effects of photon dressing, and surface effects. This will allow us to use essentially the same formalism as in the case of vacuum.

Since we want to treat the electrons as non-relativistic, we shall also restrict the energy $\hbar \Omega$ of our photons from above

$$\hbar \Omega \ll mc^2.$$  \hfill (2)

We shall also assume, trivially, that the Fermi energy of the electron gas (imagining typical metal density) satisfies

$$E_F \ll mc^2,$$  \hfill (3)

obviously satisfied in practice.
The starting point is the non-relativistic hamiltonian for electrons interacting with electromagnetic field in the Coulomb gauge

\[
H = \int d^3x \Psi^\dagger(x) \left( \frac{1}{2m} \left( \tilde{p} + \frac{e}{c} \tilde{A}(x) \right)^2 - \frac{\hbar k}{2} a^\dagger_{k\lambda} a_{k\lambda} \right) + \frac{1}{2} \int d^3x_1 d^3x_2 \Psi^\dagger(x_1) \Psi^\dagger(x_2) U(x_1 - x_2) \Psi(x_2) \Psi(x_1) ,
\]

(4)

the electron charge being \(-e\). Here \(\Psi^\dagger(x)\) creates an electron at point \(x\) (spin indices are omitted for simplicity) and \(a^\dagger_{k\lambda}\) creates a photon of wavevector \(\tilde{k}\) and polarization \(\lambda\). The \(\tilde{A}\) operator is

\[
\tilde{A}(\vec{x}) = \sum_{\vec{k}\lambda} \left( \frac{4\pi \hbar c^2}{2\Omega_{\vec{k}} V} \right)^{1/2} e^{i\vec{k}\vec{x}} e_{\vec{k}\lambda}^* (a_{\vec{k}\lambda} + a^\dagger_{-\vec{k}\lambda}) ,
\]

(5)

where \(V\) is the normalization volume. The total hamiltonian can be split into the following terms

\[
H = H_0 + H_{Coul} + H_{rad} + H_{i1} + H_{i2} ,
\]

(6)

where

\[
H_0 = \int d^3x \Psi^\dagger(x) \left( \frac{1}{2m} \tilde{p}^2 \Psi(x) \right)
\]

(7)

and

\[
H_{rad} = \sum_{\vec{k}\lambda} \hbar \Omega_{\vec{k}} a^\dagger_{\vec{k}\lambda} a_{\vec{k}\lambda}
\]

(8)

are the free fields,

\[
H_{Coul} = \frac{1}{2} \int d^3x_1 d^3x_2 \Psi^\dagger(x_1) \Psi^\dagger(x_2) U(x_1 - x_2) \Psi(x_2) \Psi(x_1)
\]

(9)

is the Coulomb interaction between the electrons and

\[
H_{i1} = \frac{e}{mc} \int d^3x \Psi^\dagger(x) \tilde{A}(x). \tilde{p} \Psi(x)
\]

(10)

and

\[
H_{i2} = \frac{e^2}{2mc^2} \int d^3x \Psi^\dagger(x) \Psi(x) \tilde{A}(x). \tilde{A}(x)
\]

(11)
are the interaction terms linear and quadratic in $A(x)$, respectively.

Now to have a non-zero $S$-matrix element $S_{fi}$ between the initial photon state containing 2 photons $k_1, k_2$ and the final state with two photons $k'_1, k'_2$, we must have in the $S$-operator $S(-\infty, +\infty)$ a term with at least four $A$ operators

$$S_{fi} = \langle \Phi_{FS} | \langle \Phi_{ph} | a_{k'_1} a_{k'_2} S(-\infty, +\infty) a_{k_1}^\dagger a_{k_2}^\dagger | \Phi_{ph} \rangle | \Phi_{FS} \rangle \, ,$$

(12)

where $| \Phi_{ph} \rangle$ and $| \Phi_{FS} \rangle$ being the photon vacuum and the ground state of the Fermi sea of electrons, respectively. The photon polarization index $\lambda$ has been absorbed into $k$. Elasticity of the process is implied by the Fermi sea being left in its ground state at the end of the process.

For photon–photon scattering in vacuum, described by the relativistic hamiltonian, where the interaction term is linear in $A$, the lowest order of perturbation theory giving nonzero contribution is the 4th \cite{1}, \cite{2}, \cite{3}. The corresponding processes are the three diagrams on the Fig.4, differing from each other by the assignment of the external legs to the vertices of the square, plus three other diagrams which differ from the former ones just by the orientation of the internal fermionic loop.

In our case, however, there are more possibilities, because the interaction term $H_{i2}$ is quadratic in $A$. The simplest scattering processes now appear in second, third and fourth order of perturbation theory, and are characterized by the respective scattering amplitudes

$$S_{fi}^{(2)} = \left( -\frac{i}{\hbar} \right)^2 \frac{1}{2!} \int_{-\infty}^{+\infty} dt_1 \int_{-\infty}^{+\infty} dt_2 \langle \Phi_{FS} | \langle \Phi_{ph} | a_{k'_1} a_{k'_2} T[H_{i2}(t_1)H_{i2}(t_2)] a_{k_1}^\dagger a_{k_2}^\dagger | \Phi_{ph} \rangle | \Phi_{FS} \rangle \, ,$$

(13)

$$S_{fi}^{(3)} = \left( -\frac{i}{\hbar} \right)^3 \frac{1}{3!} \int_{-\infty}^{+\infty} dt_1 \ldots \int_{-\infty}^{+\infty} dt_3 \langle \Phi_{FS} | \langle \Phi_{ph} | a_{k'_1} a_{k'_2} T[H_{i1}(t_1)H_{i1}(t_2)H_{i2}(t_3)] a_{k_1}^\dagger a_{k_2}^\dagger | \Phi_{ph} \rangle | \Phi_{FS} \rangle \, ,$$

(14)

$$S_{fi}^{(4)} = \left( -\frac{i}{\hbar} \right)^4 \frac{1}{4!} \int_{-\infty}^{+\infty} dt_1 \ldots \int_{-\infty}^{+\infty} dt_4 \langle \Phi_{FS} | \langle \Phi_{ph} | a_{k'_1} a_{k'_2} T[H_{i1}(t_1)H_{i1}(t_2)H_{i1}(t_3)H_{i1}(t_4)] a_{k_1}^\dagger a_{k_2}^\dagger | \Phi_{ph} \rangle | \Phi_{FS} \rangle \, ,$$

(15)

where all operators are now in the interaction picture. In the following sections we shall investigate two of the above processes, $S_{fi}^{(2)}$ and $S_{fi}^{(4)}$ in actual detail, and evaluate their
scattering amplitudes.

Since we shall finally be interested in the cross sections of the scattering processes, we recall that the usual scattering amplitudes $M_{fi}$ satisfy the relations

$$S_{fi} = \delta_{fi} + i(2\pi)^4\delta(P_f - P_i)T_{fi},$$

$$T_{fi} = \frac{1}{\sqrt{2\Omega_{k_1}V}} \ldots \frac{1}{\sqrt{2\Omega_{k_2}V}} M_{fi},$$

and that the differential scattering cross section $\frac{d\sigma}{d\Omega}$ is related to $M_{fi}$ through the equation

$$\frac{d\sigma}{d\Omega} = \frac{1}{64\pi^2c^4} \frac{|M_{fi}|^2}{(\Omega_1 + \Omega_2 - c(k_1 + k_2)\cdot \vec{n})^2},$$

where $\Omega_{1,2}$ and $\vec{k}_{1,2}$ are frequencies and wavevectors of the incident photons and $\vec{n}$ is a unit vector in the direction of the solid-angle element $d\Omega$.

### III. SECOND ORDER SCATTERING PROCESSES: PLASMONS

In this section we shall deal with the processes which result from the second order scattering amplitude $[13]$. In the end, these will be the dominant contribution to the scattering, due to the smallness of the fine structure constant $\alpha = \frac{e^2}{\hbar c} = \frac{1}{137}$. Substituting for $H_{i2}$ from (11) and using the fact that the $\Psi$ and $\vec{A}$ operators commute we get

$$S_{fi}^{(2)} = \left( \frac{-i}{\hbar} \right)^2 \frac{1}{2!} \left( \frac{e^2}{2mc^2} \right)^2 \int d^4x_1 \int d^4x_2 \langle \Phi_{FS} | T[\Psi_{i}^\dagger(x_1)\Psi_{i}(x_1)\Psi_{i}^\dagger(x_2)\Psi_{i}(x_2)] | \Phi_{FS} \rangle$$

$$\times \langle \Phi_{ph} | a^\dagger_{k_1} a^\dagger_{k_2} T[(\vec{A}(x_1)\cdot \vec{A}(x_1))(\vec{A}(x_2)\cdot \vec{A}(x_2))] a_{k_1} a_{k_2} | \Phi_{ph} \rangle.$$

Using Wick’s theorem, we find that there are six diagrams, three of which are shown on Fig. 2. The three remaining diagrams differ from these just by interchange of the vertices and thus yield exactly the same contribution. Diagrams 1 and 2 of Fig. 3 can be regarded as processes where the 4-momentum transfer between the photons is accomplished via exchange of an electron-hole pair. Diagram 3 describes instead the creation of an electron-hole pair by two-photon absorption and subsequent decay of the pair into two final photons.
Performing the usual algebra and introducing the electron Green’s functions $G^0$ we get for $M_{fi}^{(2)}$ from the bare diagrams on Fig.2 (including the factor of 2 from other 3 diagrams which just cancels the $\frac{1}{2!}$ factor)

$$M_{fi}^{(2)} = \left(-\frac{i}{\hbar}\right)^2 \left(\frac{e^2}{2mc^2}\right)^2 \left(4\pi\hbar c^2\right)^2 \sum_{\text{diagrams}} (\vec{\epsilon}_{i1}, \vec{\epsilon}_{i2}) (\vec{\epsilon}_{i3}, \vec{\epsilon}_{i4}) \frac{(-2i)}{(2\pi)^4} \int d^4k G^0(k)G^0(k + p_i)$$

$$= -\frac{1}{\hbar} \left(\frac{e^2}{2mc^2}\right)^2 \left(4\pi\hbar c^2\right)^2 \sum_{\text{diagrams}} (\vec{\epsilon}_{i1}, \vec{\epsilon}_{i2}) (\vec{\epsilon}_{i3}, \vec{\epsilon}_{i4}) \Pi^0(p_i).$$

(20)

In the last expression we have introduced the usual Lindhard complex polarizability function $\Pi^0(p_i)$ of the free electron gas [7], with $p_i$ for the i-th diagram determined by the 4-momentum conservation in the vertex. The unit polarization vectors of the photons incident with the vertex 1, resp. 2 of the i-th diagram have been denoted as $\vec{\epsilon}_{i1}, \vec{\epsilon}_{i2}$ and $\vec{\epsilon}_{i3}, \vec{\epsilon}_{i4}$, respectively.

To proceed further, we notice an important difference between the interaction hamiltonians $H_{i1}$ and $H_{i2}$. In a homogeneous medium, the electron-hole pair created by a photon through $H_{i1}$ is transverse and therefore cannot decay into a longitudinal Coulomb interaction. Hence, the process of Fig.3 has zero amplitude. If however, as in the present case (Fig.2), the electron-hole pairs are created by $H_{i2}$, this selection rule is absent, and there is nothing to prevent decay of the pair and a subsequent creation of another one due to Coulomb interaction. Therefore we must renormalize the bare diagrams of Fig.2. Within the random phase approximation (RPA) this is done by summing the whole "bubble" series of Fig.4, which we recognize as the familiar plasmon series [8]. The renormalized diagrams 1 and 2 can now be interpreted as photons interacting with each other via an exchange of virtual plasmons. The diagram 3 then corresponds to absorption of two photons with creation of an intermediate plasmon, plus subsequent decay of the plasmon into the two final photons.

As is well known, exact summation of the plasmon series is equivalent to replacing the bare polarizability $\Pi^0(p_i)$ by the screened polarizability

$$\Pi(p_i) = \frac{\Pi^0(p_i)}{1 - U_0(\vec{p}_i)\Pi^0(p_i)},$$

(21)
where

$$U_0(q) = \frac{4\pi e^2}{q^2}$$

is the bare Coulomb interaction. Our final result for the scattering amplitude then reads

$$M^{(2)}_{fi} = -\frac{1}{\hbar} \frac{e^2}{2mc^2} (4\pi \hbar c^2)^2 \sum_{\text{diagrams}} (\vec{\epsilon}_{i1} \cdot \vec{\epsilon}_{i2}) (\vec{\epsilon}_{i3} \cdot \vec{\epsilon}_{i4}) \frac{\Pi^0(p_i)}{1 - U_0(\vec{p}_i)\Pi^0(p_i)}. \quad (22)$$

Because the Lindhard function $\Pi^0(p)$ is itself a complicated function of the 4-momentum transfer $p$, the scattering cross section resulting from the last expression will also depend in an intricate way on the frequencies of the incident photons as well as on the geometry of the situation, which is determined by the wavevectors and polarizations of all the photons involved. In the rest of this section, we illustrate just a few particular cases.

First of all, we notice that if the 4-momentum transfer $p_i$ is such that the denominator $1 - U_0(\vec{p}_i)\Pi^0(p_i)$ becomes zero, the scattering amplitude (22) diverges. This corresponds to an excitation of a real, instead of a virtual, plasmon. This unphysical divergence, however, is just a consequence of the random phase approximation, which we have used for simplicity. Summing a larger class of diagrams and dressing the plasmon would cause the pole to move away from the real axis, and thus remove the divergence. Nevertheless, we still expect the scattering cross section in a realistic metal to be considerably enhanced due to this ”plasmon resonance”.

Plasmon resonances can be hit as a function of scattering geometry. The result (22) is valid for a generic geometry. For illustration, we have investigated simple planar scattering pictorially described in Fig.5. For simplicity, we further restrict the two incident photons to have equal energies and perpendicular wavevectors $\vec{k}_1 \perp \vec{k}_2 (\theta = \pi/2)$. The wavevectors of all four photons involved are therefore assumed to lie in the same plane, the polarization vectors being perpendicular to the latter. On Fig.6 we show the dependence of $\frac{d\sigma}{d\Omega}$ on the angle $\beta$ for $\hbar\Omega_1 = \hbar\Omega_2 = 10$ eV. The density of the electron gas was taken to be that of metallic potassium, with $k_F = 0.73 \times 10^8$ cm$^{-1}$, corresponding to $E_F = 2.03$ eV. The corresponding plasma frequency is $\hbar\omega_p \sim 4$ eV, well below $\hbar\Omega_1, \hbar\Omega_2$, as required. One can see sharp peaks corresponding to the plasmon resonance. The peaks occur for angles $\beta$ such
that the 4-momentum of the scattered photon, determined by conservation rules, exactly fits the requirement for excitation of a real plasmon in one of the diagrams.

This calculation shows that, at least away from the peaks, the differential cross section for ultraviolet photons is exceedingly small, of the order of $10^{-36}$ cm$^2$. It is therefore of interest to ask what the dependence on the photon frequencies will be, and particularly whether the cross section could get larger in some other regime. In order to illustrate the frequency dependence, we consider the particular case of scattering of two incident photons with opposite wavevectors (Fig. 5, $\theta = \pi$), assuming again all the photons involved to be polarized perpendicularly to the plane in which the wavevectors lie. In this case, in the diagram 3 of Fig. 2 the 3–momentum transfer is zero, and this diagram does not contribute at all. In diagrams 1 and 2 the energy transfer is zero, and therefore no plasmon resonance is possible. It is easy now to find the limiting forms of $\frac{d\sigma}{d\Omega}$ for cases $k \ll k_F$ (low frequencies) and $k \gg k_F$ (high frequencies).

For the case $k \ll k_F$, we expand $\Pi^0(p_i)/[1 - U_0(p_i)\Pi^0(p_i)]$ in (22) in powers of $|\vec{p}_i|$, and keep just the first term, which is of order $|\vec{p}_i|^2$. The resulting differential cross section is

$$\frac{d\sigma}{d\Omega} = \alpha^2 \frac{E_F}{2mc^2} \frac{k}{k_F^2} = \alpha^2 \frac{E_F}{2mc^2} \frac{\Omega^2}{c^2k_F^4}. \quad (23)$$

We notice that this result is independent of the angle $\beta$. For $\hbar\Omega = 10$ eV, it is 24 orders of magnitude larger than the bare vacuum process (Fig. 4), whose total integrated cross section in this regime is known to be [3]

$$\sigma = 0.03\alpha^2 r_e^2 \left(\frac{\hbar\Omega}{mc^2}\right)^6, \quad \hbar\Omega \ll 2mc^2,$$

where $r_e = e^2/mc^2$ is the classical electron radius.

The opposite limit $k \gg k_F$ is applicable in the range $\hbar c k_F \ll \hbar \Omega \ll mc^2$. For $k$ not too large, and the angle $\beta$ not too small, or too close to $\pi$, we expand the Lindhard function $\Pi^0(\vec{p}_i, \omega_i)$ in (22) in powers of $1/|\vec{p}_i|$ and keep just the first term, which is of order $1/|\vec{p}_i|^2$.

For the differential cross section we then obtain

$$\frac{d\sigma}{d\Omega} = \frac{4}{9\pi^2} \alpha^4 \frac{E_F}{2mc^2} \frac{k}{k_F^2} \frac{1}{\sin^4 \beta} k_F^{-2} = \frac{4\alpha^4}{9\pi^2} \frac{E_F}{2mc^2} \frac{1}{\sin^4 \beta} \frac{c^6 k_F^4}{\Omega^6}. \quad (24)$$
Fig. 7 shows a log–log plot of $\frac{d\sigma}{d\Omega}$ versus photon frequency $\Omega$ for $\beta = \pi/2$, together with the asymptotic dependences (23) and (24). We see that the differential cross section has a maximum for frequencies corresponding to the photon wavevector $k \sim k_F$, where, with the present parameters, it is of order $10^{-32} cm^2$. This is still a very small value, in fact somewhat smaller than the maximal photon–photon scattering cross section in vacuum. However, for photon frequencies in the ultraviolet and soft X–ray range, up to several keV’s, this cross section is still about 15 orders of magnitude larger than that of the bare vacuum process (Fig. 4). This is what may make it easier to measure in the end.

So far we have dealt with the 2nd order scattering processes only. The 3rd order scattering amplitude (14) gives rise to ”triangle” diagrams, like that on Fig. 8. As we see, in these diagrams one also encounters the plasmon series, since one of the vertices corresponds to the interaction term $H_{12}$. We expect the contribution of these diagrams to lie somewhere between that of the 2nd order processes, which we have investigated, and the 4th order ones, which we will discuss next. We shall not evaluate any of these 3rd order processes explicitly.

IV. HIGHER ORDER PHOTON–PHOTON SCATTERING PROCESSES: THE FOURTH–ORDER (SQUARE) DIAGRAMS

The straightforward second–order calculation of the previous section contains the main result of this paper, for the photon–photon elastic cross section in an electron gas. However, it is found, that for $\Omega \gg ck_F$, the cross section has a very fast falloff with frequency, like $\Omega^{-6}$, much faster than the $\Omega^{-2}$ behaviour known for fourth–order processes [3]. In fact, it can be anticipated, that higher order processes should become increasingly important as frequency increases. Therefore, it is at least in principle possible that third or fourth–order processes could become important when $\Omega$ is both very large with respect to $ck_F$, while still very small relative to $mc^2$ (the optimal photon frequency for vacuum electron–positron processes). For this reason, we have undertaken the much heavier task of calculating the fourth–order scattering processes for two photons in the EG. A second reason for doing
this, is that this calculation will permit a better formal comparison with photon–photon scattering in vacuum, which is strictly fourth–order.

We now consider the fourth–order scattering amplitude (15), which is the non-relativistic analog of the vacuum processes with the vertex corresponding now to the interaction term \( H_{i1} \).

Substituting for \( H_{i1} \) from (10) we get

\[
S_{fi}^{(4)} = \left( -\frac{i}{\hbar} \right)^4 \frac{1}{4!} \left( \frac{-i\hbar c}{mc} \right)^4 \int dx_1 \ldots \\
\int dx_4 \left( \Phi_{ph} | a_{k_1}^* a_{k_2}^* T[(A(x_1) \cdot \nabla_1) \ldots (A(x_4) \cdot \nabla_4)] a_{k_1}^+ a_{k_2}^+ | \Phi_{ph} \right) \\
\times \langle \Phi_{FS} | T[\Psi^\dagger(x_1) \Psi(x_1) \ldots \Psi^\dagger(x_4) \Psi(x_4)] | \Phi_{FS} \rangle, \tag{25}
\]

where each of the gradients \( \nabla_i \) acts only on \( \Psi(x_i) \). Evaluating this expression in the usual way, using Wick’s theorem, we arrive at the already mentioned six diagrams (Fig.1), corresponding to the six possibilities for contraction of the \( \Psi \) operators.

Making use of

\[
\nabla_\vec{x} G^0(x, y) = (2\pi)^{-4} \int d^4 p (ip_0) e^{ip(x-y)} G^0(p), \tag{26}
\]

and of relations (13) and (17), and collecting together all the numerical factors we get finally, omitting all details, an expression for \( M_{fi}^{(4)} \)

\[
M_{fi}^{(4)} = \frac{2i\hbar^2}{\pi^2} \left( \frac{e}{m} \right)^4 \sum_{\text{diagrams}} I_i. \tag{27}
\]

We have denoted as \( I_i \) the integral corresponding to the \( i \)-th diagram

\[
I_i = \int d^4 p (\vec{e}_{i1} \cdot \vec{p}_{i1}) \ldots (\vec{e}_{i4} \cdot \vec{p}_{i4}) G^0(p_{i1})G^0(p_{i2})G^0(p_{i3})G^0(p_{i4}), \tag{28}
\]

where \( \vec{e}_{in} \) is the photon polarization unit vector in \( i \)-th diagram, vertex \( n \).

Now since each electron Green’s function \( G^0 \) consists of two parts corresponding to electrons and holes, respectively, each of the above integrals splits into 16 terms. Each of these will contain a product of 4 step functions \( \Theta(\pm(\vec{p}_j \mid -k_F)) \), which defines an integration region that is an intersection of interiors and/or exteriors of four mutually displaced Fermi
spheres. For general direction and magnitudes of photon wavevectors, the integration over such region would be quite intricate. For the sake of simplicity, we have not attempted to evaluate all integrals for a general situation. In the Appendix we present details of a particular calculation for the special geometry of forward scattering of two photons with opposite wavevectors (i.e. two initial photons with wavevectors $\vec{k}, -\vec{k}$ scatter into the same two final photons), where the integrations turn out to be particularly easy. We shall just quote here the final results of this rather tedious calculation, where approximations have also been made based on relations (2) and (3).

We have considered two limiting cases for the photon wavevector. First we take the case $k \ll k_F$, which applies very well for ultraviolet photon energies $\hbar\Omega \ll 1$ keV. We have obtained the result, Eq. (67), which, after passing from the rescaled variables (used throughout the appendix) back to the original ones, reads

$$\sum_{\text{diagrams}} I_i = \left(\frac{2m}{\hbar}\right)^3 k_F^3 \frac{8\pi i}{3} B(\phi_1, \phi_2, \phi'_1, \phi'_2) \left(\frac{k}{k_F}\right)^3 \left(\frac{\hbar k_F^2}{2m}\right)^4 \frac{1}{\Omega^4},$$

(29)

where $k$ and $\Omega$ are the wavevector and the frequency of the photon. The function $B(\phi_1, \phi_2, \phi'_1, \phi'_2)$ is a geometrical factor, which accounts for the polarization of the photons, and is defined in equation (68) in the Appendix.

Substituting for $\Omega$ the bare photon frequency $\Omega = ck$, we get for $M_{fi}^{(4)}$

$$M_{fi}^{(4)} = -\frac{256B}{3\pi} \alpha^2 \left(\frac{E_F}{2mc^2}\right)^{3/2} \frac{c^2 \hbar^2}{\Omega} E_F^2 k_F^{-1}.$$  

(30)

The corresponding fourth–order differential scattering cross section $\frac{d\sigma}{d\omega}$ is then finally, according to (18)

$$\frac{d\sigma}{d\omega} = \frac{32B^2}{9\pi^4} \alpha^4 \left(\frac{E_F}{mc^2}\right)^3 \left(\frac{E_F}{\hbar\Omega}\right)^4 k_F^{-2}, \quad \omega_p < \sim \Omega \ll ck_F.$$  

(31)

To get an order of magnitude for $\frac{d\sigma}{d\omega}$ we substitute in the last expression the values typical for metals, i.e. $k_F \sim 10^8 cm^{-1}$ and $E_F \sim 5$ eV. With $\hbar\Omega \sim E_F$, we obtain

$$\frac{d\sigma}{d\omega} \sim (4 \times 10^{-2}) \cdot (3 \times 10^{-9}) \cdot (10^{-5})^3 \cdot 1 \cdot (10^8)^{-2} \sim 10^{-41} cm^2.$$  

(32)
The second limiting case we have investigated is the case of photon wavevectors falling above $2k_F$, which means X-ray photons with energies $h\Omega \sim 1$ keV. The sum over the diagrams is given by (70), which translated back to true variables reads

$$
\sum_{\text{diagrams}} I_i = \left( \frac{2m}{\hbar} \right)^3 k_F \frac{-64\pi i}{15} B(\phi_1, \phi_2, \phi'_1, \phi'_2) \left( \frac{k}{k_F} \right)^4 \left( \frac{\hbar k^2}{2m} \right)^4 \frac{1}{\Omega^4}.
$$

(33)

Substituting again the bare photon frequency for $\Omega$ we obtain the scattering amplitude

$$
M^{(4)}_{fi} = \frac{512B}{15\pi} \alpha^2 \left( \frac{E_F}{mc^2} \right)^2 c^2 \hbar^{-1} E_F k_F^{-1},
$$

(34)

and we notice that this does not depend on $\Omega$. The differential scattering cross section then equals

$$
\frac{d\sigma}{d\Omega} = \left( \frac{32B}{15\pi^2} \right)^2 \alpha^4 \left( \frac{E_F}{mc^2} \right)^4 c^2 \hbar^{-1} E_F k_F^{-2}, \quad 2\hbar c k_F < h\Omega \ll mc^2.
$$

(35)

We estimate the order of magnitude of $\frac{d\sigma}{d\Omega}$ for $k \sim 2k_F$ and obtain

$$
\frac{d\sigma}{d\Omega} \sim (4 \times 10^{-2}) \cdot (3 \times 10^{-9}) \cdot (10^{-5})^4 \cdot (10^{-3})^2 \cdot (10^8 \text{cm}^{-1})^{-2} \sim 10^{-52} \text{cm}^2.
$$

(36)

Equations (31) and (35) represent the main results of this section. We can now compare them with their equivalent results in vacuum, namely

$$
\sigma = 0.03 \alpha^2 r_e^2 \left( \frac{h\Omega}{mc^2} \right)^6, \quad h\Omega \ll 2mc^2
$$

(37)

$$
\sigma = 4.7 \alpha^4 \left( \frac{c}{\Omega} \right)^2, \quad h\Omega \gg 2mc^2,
$$

(38)

where $\sigma$ is the total integrated cross section for unpolarized photons and $r_e = e^2/mc^2$ is the classical electron radius. We note, first of all, that the $\alpha^2$ dependence at low frequencies is the same as in our low frequency second–order result of Section 3, Eq.(23), while the high frequency $\alpha^4$ behaviour is the same as in both the second–order result of Eq.(24), and in the present fourth–order results (31) and (35). Secondly, we see that the $\Omega^{-2}$ frequency dependence (assuming $\Omega_1 = \Omega_2 = \Omega$) is the same for both 4th order in vacuum and 4th order in the EG, provided the high-frequency regime is reached in each case. Numerically, however, the coefficient in front of $\Omega^{-2}$ is much smaller in the EG case. At lower frequencies,
instead, we find a new regime in the EG case, where the 4th order cross section falls off like $\Omega^{-4}$, Eq.(31). This regime does not exist in vacuum, and is clearly due to the presence of a Fermi surface in the electron gas problem.

We can now try an overall graphical comparison of all these results. This is sketched in Fig.9. We see that the 2nd order processes in the EG are dominant for photon energies in the range from $\omega_P$ up to $\sim 10^4 \div 10^5$ eV. For X-ray photons of $10^5$ eV the relativistic vacuum processes become important and dominate at all higher photon frequencies. Hence, the 4th order EG contributions are always negligible, and by a huge factor, in spite of their slower falloff with photon energy. It is reasonable, therefore, to assume without proof that similar conclusions will apply to the third order processes, which we have accordingly ignored.

V. DISCUSSION, AND POSSIBLE EXPERIMENTAL VERIFICATION.

We have considered the problem of elastic ultraviolet and X-ray photon-photon scattering inside a solid, idealized as a free electron gas. We find that the presence of the electron gas should give rise to new scattering processes, much more important than those present in vacuum. There, only 4th order processes, important for $\gamma$-ray photons in the MeV range, are operative. In the nonrelativistic electron gas, instead, the existence of two-photon vertices introduces large second order processes, which are most efficient for photon wavevector roughly equal to the Fermi wavevector of the metal. We also predict important plasmon resonances to take place, and an angular dependence different from that of the vacuum processes.

We have also recalculated the 4th order processes in the EG. We find that these are themselves different from those in vacuum, mostly due to the breakdown of electron-hole symmetry, and their importance increases as the photon frequency decreases. However, the plasma frequency $\omega_P$ represents in practice a lower bound for the frequency of a photon, if it should penetrate inside a metal, and we have found the 4th order processes to be negligible for frequencies higher than $\omega_P$. The overall situation is summarized by Fig.9, which also
gives an order of magnitude of cross sections for a metal such as potassium, chosen because of its low plasma frequency.

Let us now briefly consider the possibilities for experimental verification of our calculated electron-mediated photon-photon elastic scattering processes. We shall discuss two cases, both extremely idealized. In the first we consider the use of a powerful pulsed laser source, operating in the ultraviolet region, $\hbar \Omega \sim 10 \text{ eV}$. The second case will be that of an X-ray in 1 keV range, such as that, which can be obtained from a synchrotron source (plus undulators).

In order to estimate the scattering cross section for the first case, we refer to Fig. 3. Making use of a plasmon resonance, we can expect $\frac{\Delta n}{\Delta \Omega} \sim 10^{-35} \text{ cm}^2$ for a metal. One could think, for example, of a crossed-beam experiment. Two laser beams are focused on a small area and penetrate into the metal and the scattered radiation is picked up by a detector. Without worrying about problems of power dissipation, let us think of one laser pulse per second, of a duration of $T$ picoseconds and a peak power of $I \times 10^{10}$ Watts. The pulse contains an energy of $\sim I T \times 10^{-2} \text{ J}$, which corresponds to $\sim I T \times 10^{16}$ photons with energy 10 eV. The spatial length of the pulse is $3 T \times 10^{-2} \text{ cm} = 3 T \times 10^6 \text{ Å}$. Imagining a penetration depth of $\sim 200 \text{ Å}$, the number of photons inside the metal is $I \times 10^{12}$. The total scattering cross section, as seen by another photon, is therefore $\sim I \times 10^{-23} \text{ cm}^2$. If the second beam, having the same parameters as the first one, is focused on the area of $(3 \mu \text{m})^2 \sim 10^{-7} \text{ cm}^2$, the number of collisions during the pulse is $\frac{I \times 10^{-23} \text{ cm}^2}{10^{-7} \text{ cm}^2} \times I \times T \times 10^{16} \sim I^2 T$. For a peak power of, e.g., $10^{10}$ W ($I = 1$) and a pulse duration of 1 ps ($T = 1$) we can expect about one scattered photon per unit solid angle per second. This flux should be, presumably, detectable with present experimental techniques.

In the case of a synchrotron source, one can work in the soft X-ray region, where the 2nd order scattering cross section has a maximum. According to Fig. 4, this occurs for photon energies $\hbar \Omega \sim \hbar c k_F$ in the keV range, where one has $\frac{\Delta n}{\Delta \Omega} \sim 10^{-32} \text{ cm}^2$. The penetration depth in the metal in this region might be $\sim 10^4 \text{ Å}$. To be more specific, we consider a beam of 1 keV radiation, consisting of $10^9$ pulses per second, with pulse duration 100 ps and peak photon flux $10^{20} \text{ photon s}^{-1}$. If the cross section of the beam is taken $\sim 10^{-7} \text{ cm}^2$, the
resulting number of scattered photons is again \( \sim 1 \) photon per second and unit solid angle, which is similar to the crossed UV laser case, and should have no sample burning problems.

In all cases, it is clear that, apart from the products of photon-photon collisions (which we are after) there will be a background of photons scattered by other entities due to different mechanisms. There will be, for example, single-photon processes, like photon scattering from free electrons. The order of magnitude of the corresponding cross section can be estimated from classical Thomson formula to be of order \( d\sigma \sim \sigma_e \sim 10^{-25}\text{cm}^2 \), and is in general larger than the photon-photon cross section, at least for reasonable intensities. These single-photon processes, however, should be distinguishable from the two-photon processes of our interest, either by requiring photon-photon coincidence, or by making use of either the quadratic intensity dependence of the photon-photon events, or of their polarization dependence \(^{22}\).

A subtler complication is that of additional contribution of interband transitions to two-photon processes. Since interband transitions are vertical, the intermediate states in the scattering process can be real ones, at particular photon frequencies. Care should be taken, therefore, to stay away from these frequencies as much as possible. As for inelastic processes, which probably represent a big problem, one may perhaps think of using the typical elastic polarization dependence obtained in Eq. \(^{22}\), to subtract them out.

Yet another possible route, although admittedly very speculative, could be studying the subtle changes in the interference pattern which one can expect to take place at very high photon intensity in a classical interference experiment. The classical theory of interference \(^{9}\) is of course based on the superposition principle, i.e. on photon-photon scattering being exactly zero. If that is no longer true, one can expect that correlation effects building up between the photons should in principle change intensities in the interference pattern. De Martini et al. \(^{10}\), for example, have recently shown how random selection of polarization in front of the interferometer presents precisely a realization of this kind of effect. In that case, when the beams are very intense, photons become correlated with one another through the random polarizer. As a result, their distribution tends to become Bose-like, which can be imagined as "trains" of photons going down one or the other slit separately. This has been
shown to weaken the interference pattern in a characteristic and measurable manner.

In principle, when the beam is sufficiently intense and the “slits” are metallic (e.g., a half-silvered mirror), a similar kind of modification of the interference pattern should be expected even without the random polarizer. Our estimates of the cross section could in principle be used to evaluate the threshold value of the intensity in a well-defined interference geometry.

A general remark may be in order, before closing this paper. As things stand, there are two elements which suggest that our calculation might remain, at least for while, of purely academic interest. The first element is that our calculated photon-photon cross sections are still very small. Detection of these processes may require a nontrivial experimental effort. The second point is that detection of our proposed process does not in itself provide new basic information, on either the photon, or the system.

Our viewpoint on these issues is open. Without embarking in a discussion of why one should or should not try to measure the process we calculated, we have simply meant to point out, for future record, that there are new photon-photon processes, which were not discussed so far, and which are in principle of measurable intensity.

APPENDIX: DETAILS OF FOURTH-ORDER CALCULATION.

To avoid problems with ill-defined expressions we shall evaluate $M_{j_i}^{(4)}$ for the case of scattering at a small angle, i.e. $\vec{k}_1 = \vec{k}$, $\vec{k}_2 = -\vec{k}$, $\vec{k}'_1 = \vec{k} + \vec{q}$, $\vec{k}'_2 = -\vec{k} - \vec{q}$, where $\vec{q}$ must be such that $|\vec{k} + \vec{q}| = |\vec{k}'|$, and then perform the limit $|\vec{q}| \to 0$ to obtain the forward scattering amplitude. Because of the transversality of the vector potential, in the limit $|\vec{q}| \to 0$ we have

$$\vec{k}.\vec{e}_1 = \vec{k}.\vec{e}_2 = \vec{k}.\vec{e}'_1 = \vec{k}.\vec{e}'_2 = 0.$$  \hfill (39)

In order to deal with dimensionless variables we rescale quantities in the integrals (28) and introduce dimensionless frequencies and wavevectors by the relations

$$\omega = \frac{\hbar k_F^2}{2m} \omega',$$  \hfill (40)
\begin{equation}
\kappa = \kappa_F \kappa'.
\end{equation}

The integrals \( I_i \) calculated with the rescaled variables \( \omega', k' \) are related to the original ones by

\begin{equation}
I_i = \left( \frac{2m}{\hbar} \right)^3 k_F I_i',
\end{equation}

and in the following the primes in all dimensionless quantities will be omitted.

As already mentioned, each of the integrals (28) splits into 16 terms corresponding to all possible combinations of electrons and holes participating in the process. Some of these terms are, however, immediately seen to be equal to zero. Apart from obvious vanishing of the terms in which all 4 internal lines correspond simultaneously to electrons, resp. holes, terms in diagrams 1 and 3 in which there is an electron and a hole in the state with the same 3-momentum, namely \( \mathbf{p}_2 = \mathbf{p}_4 \), have to vanish, too, due to the vanishing of the integration region. In the Tab. 1 we enumerate all possibly non-zero contributions to integrals \( I_1, I_2, I_3 \), corresponding respectively to the diagrams 1, 2, 3 (Fig. 10), where + sign denotes a hole and -- sign an electron.

Each of the terms has a general form

\begin{equation}
\int d^3 \mathbf{p} \int d\omega (\epsilon_{i1} \mathbf{p}_{i1}) \ldots (\epsilon_{i4} \mathbf{p}_{i4}) \frac{\Theta[\pm(1 \mid \mathbf{p}_{i1} \mid -1)]}{\omega_{i1} - \mathbf{p}_{i1}^2 \pm i\eta} \ldots \frac{\Theta[\pm(1 \mid \mathbf{p}_{i4} \mid -1)]}{\omega_{i4} - \mathbf{p}_{i4}^2 \pm i\eta},
\end{equation}

where the momenta \( \mathbf{p}_{i1}, \ldots, \mathbf{p}_{i4} \) depend on \( \mathbf{p} \) and frequencies \( \omega_{i1}, \ldots, \omega_{i4} \) depend on \( \omega \). Performing the frequency integrals using the residue theorem and passing to the limit \( |\mathbf{q}| \to 0 \) wherever this can be done in a straightforward way we get the following expressions

\begin{align}
I_{1a} + I_{1b} &= 4\pi i \int d^3 \mathbf{p} \ldots \Theta(1 \mid \mathbf{p} \mid) \Theta(1 \mid \mathbf{p} - \mathbf{k} \mid -1) \nonumber \\
&\times \frac{-4\mathbf{k} \cdot \mathbf{p} + 2k^2}{(\Omega + 2\mathbf{k} \cdot \mathbf{p} - k^2)(\Omega - 2\mathbf{k} \cdot \mathbf{p} + k^2)^2} \quad (44) \\
I_{2a} + I_{2b} &= 4\pi i \int d^3 \mathbf{p} \ldots \Theta(1 \mid \mathbf{p} \mid) \Theta(1 \mid \mathbf{p} + \mathbf{k} \mid) \Theta(1 \mid \mathbf{p} - \mathbf{k} \mid -1) \nonumber \\
&\times \frac{1}{4\mathbf{k} \cdot \mathbf{p}} \frac{1}{(\Omega - 2\mathbf{k} \cdot \mathbf{p} + k^2)^2} \quad (45) \\
I_{3a} + I_{3b} &= 4\pi i \int d^3 \mathbf{p} \ldots \Theta(1 \mid \mathbf{p} \mid) \Theta(1 \mid \mathbf{p} + \mathbf{k} \mid) \Theta(1 \mid \mathbf{p} - \mathbf{k} \mid -1) 
\end{align}
\[
\frac{1}{4k.\vec{p}} \frac{1}{\Omega - 2k.\vec{p} + k^2} \frac{1}{-\Omega - 2k.\vec{p} + k^2}
\]

(46)

\[ I_{2d} + I_{2e} = 4\pi i \int d^3\vec{p} \ldots \Theta(|\vec{p}| - 1) \Theta(1 - |\vec{p} + \vec{k}|) \Theta(|\vec{p} - \vec{k}| - 1) \]
\[
\times \frac{1}{4k.\vec{p}} \frac{1}{(\Omega + 2k.\vec{p} + k^2)^2}
\]

(47)

\[ I_{3d} + I_{3e} = 4\pi i \int d^3\vec{p} \ldots \Theta(|\vec{p}| - 1) \Theta(1 - |\vec{p} + \vec{k}|) \Theta(|\vec{p} - \vec{k}| - 1) \]
\[
\times \frac{1}{4k.\vec{p}} \frac{1}{\Omega + 2k.\vec{p} + k^2} \frac{1}{-\Omega + 2k.\vec{p} + k^2}
\]

(48)

\[ I_{2c} = -2\pi i \int d^3\vec{p} \ldots \Theta(1 - |\vec{p}|) \Theta(|\vec{p} + \vec{k}| - 1) \Theta(|\vec{p} - \vec{k}| - 1) \]
\[
\times \frac{1}{4k.\vec{p}} \left[ \frac{1}{(\Omega + 2k.\vec{p} + k^2)^2} - \frac{1}{(\Omega - 2k.\vec{p} + k^2)^2} \right]
\]

(49)

\[ I_{3c} = -2\pi i \int d^3\vec{p} \ldots \Theta(1 - |\vec{p}|) \Theta(|\vec{p} + \vec{k}| - 1) \Theta(|\vec{p} - \vec{k}| - 1) \]
\[
\times \frac{1}{2\Omega} \left[ \frac{1}{\Omega + 2k.\vec{p} + k^2} \frac{1}{\Omega - 2k.\vec{p} + k^2} - \frac{1}{\Omega - 2k.\vec{p} - k^2} \frac{1}{\Omega + 2k.\vec{p} - k^2} \right]
\]

(50)

where \( \Omega \) is the photon frequency. We have used the relations (39) and introduced the notation

\[ [\ldots] = (\vec{\epsilon}_{i1}.\vec{p}) (\vec{\epsilon}_{i2}.\vec{p}) (\vec{\epsilon}_{i3}.\vec{p}) (\vec{\epsilon}_{i4}.\vec{p}) = (\vec{\epsilon}_{1}.\vec{p}) (\vec{\epsilon}_{2}.\vec{p}) (\vec{\epsilon}'_{1}.\vec{p}) (\vec{\epsilon}'_{2}.\vec{p}) \]

(51)

in the above expressions. In several terms we have performed a shift and/or an inversion of the integration variable \( \vec{p} \). Terms 2f and 3f are identically zero since the corresponding \( \Theta \) functions have zero product.

Terms \( I_{1c} - I_{1f} \) and \( I_{2g} - I_{2n} \) require a comment. They contain products like \( \Theta(|\vec{p}|) \Theta(|\vec{p} + \vec{q}| - 1) \), which restrict the integration volume to a thin shell about a half of the surface of the Fermi sphere (see Fig.11). The volume element of the integration region can then be written as

\[ dV = \pm d\vec{S}.\vec{q} = \pm dS\vec{p}.\vec{q}. \]

At the same time these terms contain one denominator of the form

\[ (\vec{p} + \vec{q})^2 - \vec{p}^2 = 2\vec{p}.\vec{q} + \vec{q}^2 \rightarrow 2\vec{p}.\vec{q}, \]
which in the limit $|\vec{q}| \to 0$ precisely cancels the same product in the volume element. In the limit $|\vec{q}| \to 0$ therefore these terms become well defined surface integrals. Summing all the contributions from the diagrams 1, resp. 2, we obtain expressions

$$I_{1s} = I_{1c} + I_{1d} + I_{1e} + I_{1f}$$

$$= -\pi i \int_{|\vec{p}|=1} dS \left[ \frac{1}{\Omega + 2k.\vec{p} - k^2} \frac{1}{-\Omega + 2k.\vec{p} - k^2} \right]$$

$$I_{2s} = I_{2h} + I_{2i} + I_{2j} + I_{2l} + I_{2m} + I_{2n}$$

$$= -\pi i \int_{|\vec{p}|=1} dS \left[ \frac{1}{\Omega + 2k.\vec{p} + k^2} \frac{1}{-\Omega + 2k.\vec{p} + k^2} \right],$$

where the integration region is the whole surface of the Fermi sphere. Terms $I_{2g}$ and $I_{2k}$ are zero again because of vanishing of the corresponding product of $\Theta$ functions.

Now we must take into account also contributions of other 3 diagrams which differ from those on Fig. 10 by the orientation of the internal loop. Due to the absence of charge-conjugation symmetry, Furry’s theorem does not apply, and the contributions of diagrams which differ from each other by the orientation of the internal loop may not be equal. It can be easily seen that the contributions of diagrams 1 and 3 do not depend on the orientation, while in case of diagram 2 the change of the orientation is equivalent to the change of sign of the photon frequency $\Omega$. Therefore the sum of contributions of all the diagrams can be written as

$$\sum_{\text{all diagrams}} I_i = 2(I_{1a} + I_{1b}) + 2I_{1s} + 2(I_{3a} + I_{3b}) + 2I_{3c} + 2(I_{3d} + I_{3e})$$

$$+ I_{2a}(\Omega) + I_{2a}(-\Omega) + I_{2b}(\Omega) + I_{2b}(-\Omega) + I_{2c}(\Omega) + I_{2c}(-\Omega)$$

$$+ I_{2d}(\Omega) + I_{2d}(-\Omega) + I_{2e}(\Omega) + I_{2e}(-\Omega) + I_{2s}(\Omega) + I_{2s}(-\Omega).$$

At this point we shall perform some approximations. First we make use of the conditions (4), (3), which we assumed at the very beginning of our non-relativistic treatment. We write the bare photon dispersion relation $\Omega = ck$ in the rescaled variables as

$$\Omega = Ak,$$

where
\[ A = \frac{2mc}{\hbar k_F}. \]  

(56)

Substituting to this expression a typical Fermi wavevector of metals \( k_F \sim 10^8 \text{ cm}^{-1} \) we observe that

\[ A \sim 10^2 \gg 1 \]  

(57)

holds for \( A \). Actually, the last condition is just the square root of the condition (54). Moreover, the condition (52) now turns out to be equivalent to

\[ k \ll A. \]  

(58)

Because there is always a hole participating in the process, the integration variable \( \vec{p} \) must fall *inside* at least one of the 3 Fermi spheres defined by \( | \vec{p} | \leq 1 \), \( | \vec{p} + \vec{k} | \leq 1 \) and \( | \vec{p} - \vec{k} | \leq 1 \). This implies

\[ | \vec{p} | \leq 1 + k, \]  

(59)

and due to (57) and (58) also

\[ | \vec{p} | \ll A. \]  

(60)

Now in the denominators of our integrals we encounter terms like \( \Omega \pm 2\vec{k}.\vec{p} \pm k^2 = Ak \pm 2\vec{k}.\vec{p} \pm k^2 \), and the relations (58), (59) and (57) immediately suggest that we can expand the integrands in powers of \( 1/A \) and keep just the first non-zero term. Since the expression (54) is an even function of \( \Omega \) and therefore of \( A \), the expansion contains only even powers, and represents actually an expansion in powers of \( \frac{1}{A^2} = \frac{E_F}{2mc^2} \). The first non-zero term turns out to be of the order \( 1/A^4 \) in all the integrands, and neglecting terms of the order of \( 1/A^6 \) we obtain respectively

\[ 2(I_{1a} + I_{1b}) = \pi i \int d^3\vec{p}[\ldots] \Theta(1-|\vec{p}|)\Theta(|\vec{p} - \vec{k}| - 1) \frac{16(k^2 - 2\vec{k}.\vec{p})}{\Omega^4}. \]  

(61)

\[ 2(I_{3a} + I_{3b}) + (I_{2a} + I_{2b})(\Omega) + (I_{2a} + I_{2b})(-\Omega) = \]

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\[ \pi i \int d^3 \vec{p} \, \cdots \, \Theta(1 - |\vec{p}|) \Theta(1 - |\vec{p} + \vec{k}|) \Theta(|\vec{p} - \vec{k}| - 1) \frac{4}{k \cdot \vec{p}} \frac{(2\vec{k} \cdot \vec{p} - k^2)^2}{\Omega^4} \]

\[ 2(I_{3d} + I_{3e}) + (I_{2d} + I_{2e})(\Omega) + (I_{2d} + I_{2e})(-\Omega) = \]

\[ \pi i \int d^3 \vec{p} \, \cdots \, \Theta(|\vec{p}| - 1) \Theta(1 - |\vec{p} + \vec{k}|) \Theta(|\vec{p} - \vec{k}| - 1) \frac{4}{k \cdot \vec{p}} \frac{(2\vec{k} \cdot \vec{p} + k^2)^2}{\Omega^4} \]  \hspace{1cm} (62)

\[ I_C = 2I_{3e} + I_{2e}(\Omega) + I_{2e}(-\Omega) \]

\[ = \pi i \int d^3 \vec{p} \, \cdots \, \Theta(1 - |\vec{p}|) \Theta(|\vec{p} + \vec{k}| - 1) \Theta(|\vec{p} - \vec{k}| - 1) \frac{-16k^2}{\Omega^4} \]  \hspace{1cm} (63)

\[ I_S = 2I_{1s} + I_{2s}(\Omega) + I_{2s}(-\Omega) \]

\[ = \pi i \int_{|\vec{p}|=1} dS \, \cdots \, -4k^2 \frac{4k^2}{\Omega^4} (2\vec{k} \cdot \vec{p} + k^2) \]  \hspace{1cm} (64)

where we have already grouped together some terms.

Now we could, in principle, calculate the above integrals in a straightforward way and obtain the scattering amplitude for any \( k \), restricted only by the condition \((58)\). For sake of simplicity we shall do the integrations explicitly for 2 limiting cases, in which the integration region becomes particularly simple, first for \( k \ll 1 \), and then for \( 2 < k \ll A \).

The condition \( k \ll 1 \) allows us once again to pass in several terms to surface integration (in the same way as above when we were performing the limit \(|\vec{q}| \to 0\)). In the limit \( k \ll 1 \), the integration regions of the terms 1a,1b,2a,2b,3a,3b,2d,2e,3d and 3e become the same and equal to the half of the surface of the Fermi sphere \(|\vec{p}| = 1\), for which \( \vec{k} \cdot \vec{p} \leq 0 \) holds. Collecting these terms together and denoting their sum as \( I_I \) we obtain

\[ I_I = 2(I_{1a} + I_{1b}) + 2(I_{3a} + I_{3b}) + (I_{2a} + I_{2b})(\Omega) + (I_{2a} + I_{2b})(-\Omega) \]

\[ + 2(I_{3d} + I_{3e}) + (I_{2d} + I_{2e})(\Omega) + (I_{2d} + I_{2e})(-\Omega) \]

\[ = \frac{-\pi i}{\Omega^4} \int_{\vec{k} \cdot \vec{p} \leq 0} dS \, \cdots \, (8k^4 + 16k^2 \vec{k} \cdot \vec{p}) \]  \hspace{1cm} (65)

The remaining terms in \((54)\) are the volume integral \( I_C \) \((54)\) and the surface integral \( I_S \) \((55)\).

Since we are considering the limiting case \( k \ll 1 \), it is now enough to identify in the sum \( I_I + I_C + I_S \) terms which contain the leading power of \( k \). In \( I_I \) the leading term in \( k \) is of
order $k^3$. It is easily seen that the integration volume of the integral $I_C$ behaves as $k^3$ and the contribution of this term is therefore of order $k^5$. In the surface integral $I_S$ we encounter a term $\sim k^2 \vec{k}.\vec{p}$, which, however, vanishes after the integration over the whole surface of the Fermi sphere and the remaining term contributes as $k^4$. In the limit $k \ll 1$ therefore the dominant contribution to the scattering amplitude comes from the leading term in (66), which is readily evaluated as

$$\sum_{all\, diagrams} I_i = \frac{8\pi i}{3} \frac{k^3}{\Omega^4} B(\phi_1, \phi_2, \phi'_1, \phi'_2).$$ \hspace{1cm} (67)$$

In the last expression we introduced a function

$$B(\phi_1, \phi_2, \phi'_1, \phi'_2) = \int_0^{2\pi} d\phi \cos(\phi - \phi_1) \cos(\phi - \phi_2) \cos(\phi - \phi'_1) \cos(\phi - \phi'_2) = \frac{\pi}{4} \times$$

$$\times \left[ \cos(\phi_1 + \phi_2 - \phi'_1 - \phi'_2) + \cos(\phi_1 - \phi_2 + \phi'_1 - \phi'_2) + \cos(\phi_1 - \phi_2 - \phi'_1 + \phi'_2) \right]. \hspace{1cm} (68)$$

which represents a factor taking into account the polarization of the photons, and $\phi_1, \phi_2, \phi'_1, \phi'_2$ are direction angles of the unit polarization vectors of the photons.

In another limiting case, $2 < k \ll A$, the Fermi spheres are displaced from one another by a distance larger than 2 radii, and therefore do not intersect anymore. The integration region of integrals $I_{2a} + I_{2b}$ and $I_{3a} + I_{3b}$ becomes empty and that of the remaining volume integrals becomes the whole interior of one of the spheres. Denoting the sum of all volume integrals as $I_V$ we obtain

$$I_V = \frac{\pi i}{\Omega^4} \int d^3\vec{p} \cdots [\Theta(1 - |\vec{p}|) \left[ -32k.\vec{p} + \frac{4(2\vec{k}.\vec{p} - k^2)^2}{k.(\vec{p} - k)} \right] = \frac{4\pi i}{\Omega^4} \times$$

$$\times B(\phi_1, \phi_2, \phi'_1, \phi'_2) \left\{ \frac{k^4}{45}(-33 + 40k^2 - 15k^4) + \frac{1}{6}(k^3 - 3k^5 + 3k^7 - k^9) \ln \frac{k}{k + 1} \right\}. \hspace{1cm} (69)$$

Apart from this, there is also the surface integral $I_S$ (55), which equals

$$I_S = -\frac{64\pi i k^4}{15 \Omega^4} B(\phi_1, \phi_2, \phi'_1, \phi'_2). \hspace{1cm} (70)$$

Now for large $k$, the leading power of $k$ in $I_V$ is $k^2$. The dominant contribution to the scattering amplitude will therefore come from $I_S$, which behaves as $k^4$. However, on the side of large $k$ we are still restricted by the condition (58), and the validity of expressions
(59) and (70) is thus limited, say, to $2 < k < 10$. For this interval of $k$, we have numerically checked the relative contributions of $I_V$ and $I_S$, and really found $I_V$ to be negligible. The final result is therefore given by (70).

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REFERENCES

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1 R. Karplus, M. Neuman, Phys. Rev. 83, 776 (1951).

2 B. De Tollis, Nuovo Cimento 32, 757 (1964); 35, 1182 (1965).

3 *Relativistic Quantum Theory, Part 2*, E. M. Lifshitz, L. P. Pitaevskii, Course of Theoretical Physics, Vol. 4 (Pergamon Press, Oxford 1973)

4 A. F. Garito, Mol. Cryst. & Liq. Cryst. (GB) 106, 219 (1984)

5 Chang-qin Wu, Xin Sun, Phys. Rev. B, 41, 12485 (1990).

6 Weikang Wu, Phys. Rev. Lett., 61, 1119, 1988.

7 A. L. Fetter, J. D. Walecka, *Quantum Theory of Many–Particle Systems* (McGraw–Hill, New York 1971).

8 D. Pines, *Elementary excitations in solids* (Benjamin, New York 1963).

9 R. Loudon, *The Quantum Theory of Light* (Oxford University Press, 1973).

10 F. De Martini, S. Di Fonzo, Europhysics Lett., 10 (2), 123 (1989)

11 E. Iacopini, E. Zavattini, Phys. Lett. B, 85, 151 (1979).

12 E. Iacopini, B. Smith, G. Stefanini, E. Zavattini, Il Nuovo Cimento, 61 B, 21 (1981).
FIGURES

FIG. 1. The vacuum photon–photon fourth–order scattering diagrams.

FIG. 2. Second order photon–photon scattering processes in a nonrelativistic electron gas.

FIG. 3. A diagram with zero amplitude.

FIG. 4. Screening of the bare polarizability in the second order diagram.

FIG. 5. Photon–photon planar scattering geometry (chosen for simplicity of illustration). \( \vec{k}_1, \vec{k}_2 \) incoming photons, \( \vec{k}_1', \vec{k}_2' \) outgoing photons.

FIG. 6. Differential cross section versus angle \( \beta \) for the 2nd order processes in the planar geometry of Fig. 5, and for \( \theta = \pi/2 \). The peaks are due to the plasmon resonance. Their sharpness is a consequence of the random–phase approximation, where the plasmon lifetime is infinite.

FIG. 7. A log–log plot of the calculated differential cross section versus photon frequency for the 2nd order elastic photon–photon scattering processes in an electron gas (away from plasmon resonances). Parameters chosen are representative for potassium metal.

FIG. 8. "Triangle" diagrams corresponding to the 3rd order processes.

FIG. 9. An overall graphical comparison of the photon–photon elastic cross section in an electron gas and in vacuum. In an actual solid, or liquid, both contributions will be present.

FIG. 10. Three diagrams corresponding to the 4th order processes in an electron gas for the particular case of the scattering of two incident photons with opposite wavevectors.

FIG. 11. The integration region, which is a thin outer shell of the Fermi sphere.
TABLE I. Terms giving a non-zero contribution to the diagrams 1-3. + denotes a hole, − an electron.