Coherent photovoltaic effect due to the quantum corrections

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The appearance of a steady current produced by two electromagnetic waves with frequencies ω and 2ω—the coherent photovoltaic effect—has been studied [G. M. Shmelev *et al.*, Izv. Vuzov, Fizika **28**, 84 (1985); M. V. Éntin, Fiz. Tekh. Poluprovodn. (Sov. Phys. Semicond.) (in press)]. In the frequency region $\tau_{\varphi}^{-1} \lesssim \omega \ll \tau_{p}^{-1}$, where $\tau_{\varphi(p)}$ is the phase (momentum) relaxation time, the current is found to be determined primarily by the quantum corrections to the nonlinear rf conductivity.

In the presence of two coherent light beams $\mathbf{E}(t) = 2\mathrm{Re}(\mathbf{E}_{\omega} e^{i\omega t} + \mathbf{E}_{2\omega} e^{2i\omega t})$ and $\mathbf{E}_{-\omega} = \mathbf{E}_{\omega}^{*}$ a steady electric current can appear in any conducting medium. The general expression for the nonlinear response at the frequency ω_1 in an isotropic medium is

$$\mathbf{j}_{\omega_1} = \mathbf{E}_{\omega} \alpha_{\omega_1}(E_{\omega}^2, E_{2\omega}^2, (\mathbf{E}_{\omega} \mathbf{E}_{2\omega})) + \mathbf{E}_{2\omega} \beta_{\omega_1}(E_{\omega}^2, E_{2\omega}^2 (\mathbf{E}_{\omega} \mathbf{E}_{2\omega})) \quad (1)$$

A power series expansion of the fields in $\mathbf{j}_{\omega 1}$ gives rise to various combination harmonics with the frequencies $\omega_1 = 2n\omega + m\omega$. Among them there is a zero frequency which corresponds to a steady current. In the lowest order in the electric field $\omega_1 = 0$ at $n = 1$ and $m = -2$ and the coherent photovoltaic effect is described by the expression

$$\mathbf{j} = \alpha_1 \mathbf{E}_{2\omega} \mathbf{E}_{-\omega}^2 + \alpha_2 \mathbf{E}_{-\omega} (\mathbf{E}_{-\omega} \mathbf{E}_{2\omega}). \quad (2)$$

If the field components are represented as $E_{\omega,k} = \mathscr{E}_{\omega,k} \exp(i\varphi_{\omega k})$ and $E_{2\omega,k} = \mathscr{E}_{2\omega,k} \exp(i\varphi_{2\omega k})$ where $\mathscr{E}_{\omega}$ and $\mathscr{E}_{2\omega}$ are real values, we can see from (2) that the contribution to the current from the corresponding field components is determined by the phase factor $\exp\{i(\varphi_{2\omega k} - \varphi_{\omega j} - \varphi_{\omega e})$. In the special case of linear polarization the phases of all the field components of the given frequency are the same and the contributions to the current from the terms with $\alpha_1 = |\alpha_1| \exp(i\psi_1)$, $\alpha_2 = |\alpha_2| \exp(i\psi_2)$ are proportional to $\cos(\varphi_{2\omega} - 2\varphi_{\omega} + \psi_{1,2})$. Since the phase sensitivity distinguishes the effect under consideration from the known photovoltaic effect, we call it a coherent effect. In the case of coherent photovoltaic effect the absence of an inversion center is obviously unimportant.

Let us consider the coherent photovoltaic effect in the free-carrier absorption region in the classical frequency range $\hbar\omega \ll \epsilon$, where ϵ is the electron energy. To determine the coefficients $\alpha_{1,2}$, we must solve the classical kinetic equation in the third order in the electric field.[2] If $\omega\tau_p \ll 1$

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$$\alpha_{1,2} = -\frac{2e^4}{dm^2}\int d\epsilon\nu(\epsilon)\epsilon\tau_p \frac{\partial}{\partial\epsilon}\{T_{1,2}(\epsilon)(1+\frac{2}{d}\epsilon\frac{\partial}{\partial\epsilon})\tau_p\frac{\partial f_0}{\partial\epsilon}\}, \quad (3)$$

where d is the dimensionality of the system, $\nu(\epsilon)$ is the state density,

$$T_1(\epsilon) = \tau_\epsilon(1-2i\omega\tau_\epsilon)^{-1}, \qquad T_2(\epsilon) = 2\tau_\epsilon(1+i\omega\tau_\epsilon)^{-1},$$

here τ_ϵ is the energy relaxation time. It is assumed that the action of the inelastic part of the collision integral on the isotropic part of the distribution function $\bar{f}(p)$ can be replaced by the expression $\bar{f}(p)/\tau_\epsilon$.

In the case of a degenerate electron gas in the low-frequency limit $\omega\tau_\epsilon \ll 1$ at temperatures T lower than the Debye temperature we have $j \sim \sigma_0 E_{2\omega}(eE_\omega L_\epsilon/kT)^2$, where L_ϵ is the cooling length, and σ_0 is the ohmic conductivity. In the limit $\omega\tau_\epsilon \gg 1$ the current acquires an additional factor $(\omega\tau_\epsilon)^{-1}$ in the case of scattering by phonons or it acquires factor $(\omega\tau_\epsilon)^{-1}(kT/\epsilon_F)^2$ when the momentum relaxation is governed by the impurities.

In addition to the purely classical contribution, there is another contribution to the coherent photovoltaic effect which is attributable to the quantum corrections. This contribution arises as a result of variation, in synchronism with the extracting field 2ω, of the phase of the wave function of the electron which returns to the original position as a result of the application of the field $E_{-\omega}$. This situation causes a relative increase of the quantum correction at the half period, when the electron accelerates, say, to the right, and causes its relative decrease at the other half period; i.e., a direct current is generated. A similar contribution to the coherent photovoltaic effect occurs if $E_{-\omega}$ appears as the extracting field and if the phase of the wave function is determined by the joint action of $E_{-\omega}$ and $E_{2\omega}$.

To determine α_1 and α_2, we can use the equations for the quantum corrections to the nonlinear response[3]

$$j_{\omega_1} = -\frac{2e^2D\tau_p}{\pi^2}\int d\omega_2 E(\omega_2)\int_{-\infty}^{+\infty} dt \int_0^\infty d\eta \exp\{i[t(\omega_2-\omega_1)+\eta\frac{\omega_1+\omega_2}{2}]\}$$

$$\times \int \frac{d^dq}{(2\pi)^d} \exp\{-D\int_{-\eta}^{\eta} d\eta_1 [q-\frac{e}{c}A(t-\frac{\eta_1}{2})-\frac{e}{c}A(t+\frac{\eta_1}{2})]^2 - \frac{2\eta}{\tau_\varphi}\}.$$

Here D is the diffusion coefficient of electrons, $E(\omega)$ is the Fourier harmonic of the field $E(t)$, and $A(t)$ is the vector potential of this field.

Expanding the exponential function up to the second order in A, we find

$$\alpha_{1,2} = \alpha f_{1,2}(\omega\tau_\varphi), \quad \alpha = -\frac{64}{(16\pi)^{d/2}} \frac{e^4}{\hbar^3} \tau_\varphi^2 (D\tau_\varphi)^{2-(d/2)}$$

$$\left\{ \begin{matrix} f_1(y) \\ f_2(y) \end{matrix} \right\} = y^{(d/2)-4} \times \int \frac{dx}{x^{d/2}} e^{-4x/y} \left\{ \begin{matrix} e^{2ix}\left(x + \frac{\sin 2x}{2} - \frac{2\sin^2 x}{x}\right) \\ e^{-ix}\left(\sin x + \frac{1}{3}\sin 3x - \frac{\cos x - \cos 3x}{2x}\right) \end{matrix} \right\},$$

The functions $f_1(y)$ and $f_2(y)$, which determine the frequency dependence of the effect, decrease in the high and low frequency limits:

$$f_2(y) = 4f_1(y) = \frac{2^{9-d}}{45} \Gamma\left(6 - \frac{d}{2}\right) y^2 \qquad \text{for } y \ll 1,$$

$$f_2(y) = 2f_1(t) = -i\pi/4y^3 \qquad \text{for } y \gg 1,\ d = 1,$$

$$f_2(y) = -2f_1(y) = -i\ln y/2y^3 \qquad \text{for } y \gg 1,\ d = 2,$$

$$\left. \begin{aligned} f_1(y) &= \frac{\sqrt{\pi}}{6y^{5/2}} (-19 + 13\sqrt{2})(1+i) \\ f_2(y) &= \frac{\sqrt{\pi}}{3y^{5/2}} [3(\sqrt{2}-1) + i(3-2\sqrt{2})] \end{aligned} \right\} \qquad \text{for } y \gg 1, d = 3.$$

The effect reaches a maximum value when $\omega\tau_\varphi \sim 1$, and its order of magnitude in this case is determined by α. Since generally $\tau_p \ll \tau_\varphi \ll \tau_\epsilon$, the ratio of the quantum and classical contributions to the coherent photovoltaic effect in this region is determined by the expression $(\epsilon_F \tau_p/\hbar)(\tau_\varphi/\tau_p)^2$, which is greater than unity (the temperature is assumed to be sufficiently low, so the momentum relaxes primarily on impurities). The large contribution from the quantum effects is explained by the fact that at $\omega\tau_\varphi \sim 1$ the field, which is sufficiently large for the disruption of the electron phase, is still too small to heat the electron gas. The classical component of the coherent photovoltaic effect is larger than its quantum component only at the frequencies $\omega < \tau_\varphi^{-1} \times (\hbar\epsilon_F \tau_p \tau_\epsilon)^{1/2}/kT(\tau_\varphi)^{3/2}$.

A photovoltaic current, which is proportional to the square of the alternating field and which is associated with the quantum corrections, has been detected in mesoscopic samples.[4] This purely fluctuational current is attributable to the local decrease of the symmetry. The direction of the current, for example, fluctuates from

sample to sample. In contrast with this situation, the coherent photovoltaic effect, because of the quantum corrections, does not require the symmetry to be lowered. The sign of this macroscopic effect is determined by the polarization and the phase of the light beam. The coherent photovoltaic effect requires, however, that the electromagnetic fields be mutually coherent. This mutual coherence can be achieved by doubling the frequency in a nonlinear medium.

[1]G. M. Shmelev, Nguen Hong Shon, and G. I. Tsurkan, Izv. vuzov, Fizika, **28**, 84 (1985).
[2]M. V. Énin, Fiz. Tekh. Poluprovodn. [Sov. Phys. Semicond.] (in press).
[3]B. L. Altshuler *et al.*, in: Quantum Theory of Solids, ed. by I. M. Lifshitz, Mir, Moscow, 1982.
[4]V. I. Fal'ko and D. E. Khmel'nitskiĭ, Zh. Eksp. Teor. Fiz. [Sov. Phys. JETP] (in press).

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