Second-harmonic generation in two-dimensional systems without inversion centers

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Doubling of the frequency of a light wave on reflection from a two-dimensional electron layer without an inversion center is investigated. It is shown that for normal incidence of the radiation on the two-dimensional layer, when the wavefront is parallel to the layer and the electric field in the layer is uniform, the presence of an external magnetic field **H** parallel to plane of electron motion leads to a finite nonlinear conductivity. The current, quadratic in the incident-field amplitude and varying like $\exp(-2i\omega t)$ is of the form $\mathbf{J}^{(2)} \sim [\mathbf{c}\times\mathbf{H}]\mathbf{E}^2$, where **c** is the vector of one of the nonequivalent normals to the layer, and **E** is the electric field of the wave and varies like $\exp(-i\omega t)$. The proportionality coefficient is obtained for the case of sufficiently high frequencies ω.

1. INTRODUCTION

One of the basic processes in nonlinear optics of metals is the frequency doubling of light reflected from its surface, or second-harmonic generation. Usually the nonlinear current induced in a metal by a light beam

$$\mathbf{J}^{(2)} = \beta\mathbf{E}(\nabla\mathbf{E}) + \delta(\mathbf{E}\nabla)\mathbf{E} + \gamma\nabla\mathbf{E}^2 \tag{1}$$

is not only proportional to the square of the wave amplitude but contains also derivatives with respect to the coordinates, i.e., it is nonlocal.[1] This is due to the presence of an inversion center in the symmetry group of the metal. This current vanishes in the limit of a spatially homogeneous electric field. A local limit is naturally imposed on the alternating high-frequency electric field in the case of heterostructures of various kinds, where the carrier motion is two-dimensional when the electromagnetic wave is almost normally incident on the electron layer.

Studies[2,3] of paramagnetic and cyclotron resonances in certain heterostructures based on gallium arsenide have shown the carriers to have no degeneracy in spin. This phenomenon was interpreted as the existence of a spin-orbit interaction induced by parity violation with respect to reflection in the plane of the layer. In Ref. 4 it was proposed to describe this situation phenomenologically by adding to the carrier-effective-mass Hamiltonian a spin-orbit term, previously derived[5,6] from symmetry considerations, to describe the electron bands in three-dimensional semiconductors without inversion centers:

$$\mathcal{H}_{so} = \frac{\alpha}{\hbar}[\mathbf{pc}]\boldsymbol{\sigma}, \tag{2}$$

where **p** is the electron (or hole) momentum operator, $\boldsymbol{\sigma}$ are Pauli matrices, and **c** is a unit vector normal to the layer.

If the number of electrons in such a layer is large enough, it can be regarded as a two-dimensional metal whose symmetry group has no inversion center. Just the same, however, the nonlinear response should still vanish in the local limit. Indeed, the fact that one of the normals to the layers is singled out leads to a special direction **c**, but no current can flow along **c**, since an electric field parallel to **c** cannot excite electronic transitions of transverse size-quantized motion. To obtain a finite local limit it is necessary to have a singled-out definite direction in the layer itself. It will be shown below that it suffices for this purpose to apply a constant magnetic field **H** along the layer, in which case the current is directed along the vector $\mathbf{c}\times\mathbf{H}$.

2. EFFECTIVE HAMILTONIAN OF TWO-DIMENSIONAL MOTION

It seems almost obvious that if the cyclotron frequency ω_c is much lower than the charateristic frequencies $\varepsilon_\perp/\hbar$ of the motion across the layer, a magnetic field **H** parallel to the layer influences only the motion of the electron spin. To verify this we choose a coordinate frame with axis $\hat{\mathbf{z}}\|\mathbf{c}$ and consider the Hamiltonian of three dimensional motion in a potential $W(z)$ that forms a two-dimensional quantum well:

$$\mathcal{H}_3 = \frac{1}{2m}\left(\mathbf{p}+\frac{e}{c}\mathbf{A}\right)^2 + \frac{\alpha}{\hbar}\left[\left(\mathbf{p}+\frac{e}{c}\mathbf{A}\right)\mathbf{c}\right]\boldsymbol{\sigma} + g\boldsymbol{\sigma}\mathbf{H} + W(z). \tag{3}$$

Let $\hat{\mathbf{y}}$ be a unit vector along the field **H**. Choice of the gauge

$$A_x = Hz, \quad A_y = A_z = 0$$

permits $\mathcal{H}_3$ to be represented by the sum

$$\mathcal{H}_3 = \mathcal{H}_0 + U_1 + U_2, \quad \mathcal{H}_0 = p_z^2/2m + W(z), \tag{4}$$

$$U_1 = \frac{p_y^2}{2m} + \frac{\alpha}{\hbar}p_y[\hat{\mathbf{y}}\mathbf{c}]\boldsymbol{\sigma} + g\sigma_y H,$$

$$U_2 = \frac{1}{2m}\left(p_x + \frac{eH}{c}z\right)^2 + \frac{\alpha}{\hbar}\left(p_x + \frac{eH}{c}z\right)[\hat{\mathbf{x}}\mathbf{c}]\boldsymbol{\sigma},$$

in which the operator U_2 mixes the longitudinal and transverse motions. If the condition

$$\delta = \hbar\omega_c/\varepsilon_\perp \ll 1 \tag{5}$$

is met, the Hamiltonian U_2 is a perturbation, small in the parameter δ, to the transverse motion. The longitudinal-motion Hamiltonian is determined primarily by the mean value of $\mathcal{H}$ on the wave function $\varphi_0(z)$ of the ground state in the quantum well:

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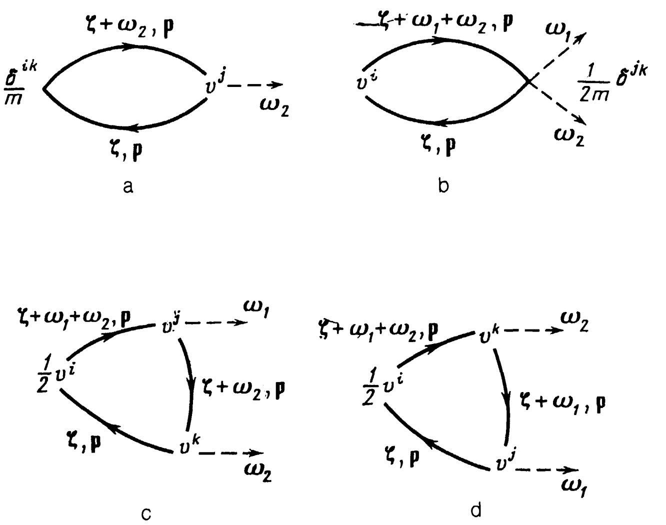

FIG. 1.

$$\mathscr{H}_2 \approx \frac{1}{2m}\left(p_x+\frac{eH}{c}z_0\right)^2+\frac{p_y^2}{2m}+g\boldsymbol{\sigma}\mathbf{H}+\frac{\alpha}{\hbar}\left[-\left(p_x+\frac{eH}{c}z_0\right)\sigma_y+p_y\sigma_x\right], \tag{6}$$

where it is taken into account that even though $z_0^2 = \langle\varphi_0|z|\varphi_0\rangle^2 \neq \langle\varphi_0|z^2|\varphi_0\rangle$, the contribution made to $\mathscr{H}_2$ to the difference of these expressions is independent of the dynamic variables of the longitudinal motion and is small in δ. The contribution of perturbation theory of second-order in U_2, which includes virtual transitions to excited states of the Hamiltonian $\mathscr{H}_0$, leads only to a small renormalization, in terms of δ, of the scalar and spinor parts of the Hamiltonian $\mathscr{H}_2$. The parameter z_0 in expression (6) is eliminated by the canonical transformation

$$U(r)=\exp(-i\mathbf{a}r), \quad \mathbf{a}=(-eHz_0/c, 0). \tag{7}$$

Here and below all the vectors are two-dimensional.

Thus, the effective-Hamiltonian of the longitudinal motion takes the form

$$\mathscr{H}=\frac{\mathbf{p}^2}{2m}+\frac{\alpha}{\hbar}[\mathbf{p}\mathbf{c}]\boldsymbol{\sigma}+g\boldsymbol{\sigma}\mathbf{H}. \tag{8}$$

3. NONLINEAR CONDUCTIVITY

The electromagnetic interaction of radiation with a two-dimensional gas is of the form

$$V=V_1+V_2, \quad V_1=\frac{e}{c}\int d\mathbf{r}\,\mathbf{v}(\mathbf{r})\mathbf{A}(\mathbf{r},t),$$
$$V_2=\frac{e^2}{2mc^2}\int d\mathbf{r}\,n(\mathbf{r})\mathbf{A}^2(\mathbf{r},t), \tag{9}$$

where $\mathbf{A}(\mathbf{r}, t)$ is the vector potential of the wave field, $n(\mathbf{r})$ the electron-density operator, and $\mathbf{v}(\mathbf{r})$ is the velocity operator, which has in this case, besides the usual scalar part, also a spin component

$$\mathbf{v}(\mathbf{r})=\frac{i\hbar}{2m}(\nabla\psi^+(\mathbf{r})\psi(\mathbf{r})-\psi^+(\mathbf{r})\nabla\psi(\mathbf{r}))+\frac{\alpha}{\hbar}\psi^+(\mathbf{r})[\mathbf{c}\boldsymbol{\sigma}]\psi(\mathbf{r}). \tag{10}$$

We confine ourselves hereafter to the case of sufficiently close incident-radiation frequencies ω, when the influence of the impurities can be neglected. The corresponding estimates will be discussed below.

The expectation value of the current operator

$$\mathbf{J}(\mathbf{r},t)=\mathbf{J}_0(\mathbf{r})+\mathbf{J}_1(\mathbf{r},t), \quad \mathbf{J}_0(\mathbf{r})=-e\mathbf{v}(\mathbf{r}),$$
$$\mathbf{J}_1(\mathbf{r},t)=-\frac{e^2}{mc}n(\mathbf{r})\mathbf{A}(\mathbf{r},t) \tag{11}$$

at the doubled frequency can be obtained by the method of Ref. 7 as the quadratic response to an alternating field. It is clear from (9) and (11) that to this end it is necessary to find the expectation value of the current $\mathbf{J}_1$ to first order perturbation theory in V_1 and the expectation value of the current $\mathbf{J}_0$ to second order in V_1. In the limit of a wave field homogeneous in space it is convenient to represent the result in the form

$$J_i^{(2)}(\Omega)=\int\sigma_{i,jk}^{(2)}(\Omega|\omega_1\omega_2)E_j(\omega_1)E_k(\omega_2)\times\delta(\Omega-\omega_1-\omega_2)d\omega_1\,d\omega_2/(2\pi)^2, \tag{12}$$

where the expression for the nonlinear conductivity $\sigma^{(2)}$ is given by a sum of diagrams (see the figure), accurate to a factor $-e^3/\omega_1\omega_2$ that appears when the vector potential is replaced by the electric field $\mathbf{E}(\omega) = (i\omega/c)\mathbf{A}(\omega)$. The solid lines corresponds to the electron Green's function

$$G_{\alpha\beta}(\zeta,\mathbf{p})=(\zeta-\mathscr{H})^{-1}=\frac{\Pi_{\alpha\beta}^{(+)}(\mathbf{p})}{\zeta-E_{(+)}(\mathbf{p})}+\frac{\Pi_{\alpha\beta}^{(-)}(\mathbf{p})}{\zeta-E_{(-)}(\mathbf{p})}, \tag{13}$$

where

$$E_{(\pm)}=\frac{\mathbf{p}^2}{2m}\pm\left(\frac{\alpha^2}{\hbar^2}\mathbf{p}^2+g^2\mathbf{H}^2+2\frac{\alpha g}{\hbar}\mathbf{p}[\mathbf{c}\mathbf{H}]\right)^{1/2} \tag{14}$$

are the energies of the two branches of the energy spectrum, and

$$\Pi^{(\pm)}=\frac{1}{2}(1\pm\mathbf{P}\boldsymbol{\sigma}), \tag{15}$$

$$\mathbf{P}(\mathbf{p})=\left(\frac{\alpha}{\hbar}[\mathbf{p}\mathbf{c}]+g\mathbf{H}\right)\Big/\left|\frac{\alpha}{\hbar}[\mathbf{p}\mathbf{c}]+g\mathbf{H}\right|$$

are the operators of projection on these branches. In the vertices are either unit operators or the velocity operators

$$\mathbf{v}=\mathbf{v}_c+\mathbf{v}_{sp}, \quad \mathbf{v}_c=\frac{\mathbf{p}}{m}, \quad \mathbf{v}_{sp}=\frac{\alpha}{\hbar}[\mathbf{c}\boldsymbol{\sigma}]. \tag{16}$$

The summation over the frequencies $\zeta = i\pi T(2n+1)+\mu$ in the triangular diagrams is carried out with allowance for the equality

$$-T\sum_\zeta(\zeta+\omega_1+\omega_2-E_a)^{-1}(\zeta+\omega_2-E_b)^{-1}(\zeta-E_c)^{-1}$$
$$=\frac{n_F(E_b)-n_F(E_c)}{(E_c-E_a+\omega_1+\omega_2)(E_c-E_b+\omega_2)}+\frac{n_F(E_b)-n_F(E_a)}{(E_a-E_c-\omega_1-\omega_2)(E_a-E_b-\omega_1)}, \tag{17}$$

where T is the temperature, assumed hereafter to be zero, μ the chemical potential, and $n_F(E)$ the Fermi occupation numbers. The energies of the three lines can take on two values $E_{(\pm)}(\mathbf{p})$. From among all the possible combinations, it is necessary to retain only those in which the corresponding occupation-number differences are not zero. This means, in particular, that on circuiting the loop a transition between different branches of the spectrum must take place at least in one vertex.

Since the same considerations hold also for diagrams *a* and *b*, and one of their vertices contains unity, their contribution vanishes by virtue of the orthogonality of the projectors $\Pi_{(+)}(\mathbf{p})$ and $\Pi_{(-)}(\mathbf{p})$.

It is necessary next to substitute for the Green's functions expression (13) and calculate the traces. All are expressed in terms of two independent variables

$$S^{ijk}_{(+-+)} = \mathrm{Sp}(v^i\Pi_{(+)}v^j\Pi_{(-)}v^k\Pi_{(+)}) = \frac{\alpha^2}{\hbar^2}\left(\frac{p^i}{m}+\frac{\alpha}{\hbar}[\mathbf{cP}]^i\right)(\delta^{jk}-[\mathbf{cP}]^j[\mathbf{cP}]^k),$$

$$S^{ijk}_{(-+-)} = \mathrm{Sp}(v^i\Pi_{(-)}v^j\Pi_{(+)}v^k\Pi_{(-)}) = \frac{\alpha^2}{\hbar^2}\left(\frac{p^i}{m}-\frac{\alpha}{\hbar}[\mathbf{cP}]^i\right)(\delta^{jk}-[\mathbf{cP}]^j[\mathbf{cP}]^k). \tag{18}$$

As a result, the expressions for diagram *c* take the form

$$\int\frac{d^2p}{(2\pi)^2}\Big\{S^{jki}_{(+-+)}\frac{n^+-n^-}{(-\Delta+\omega_1+\omega_2)(-\Delta+\omega_2)} + S^{ijk}_{(+-+)}\frac{n^--n^+}{(\omega_1-\Delta)(\omega_2+\Delta)}$$
$$+S^{kij}_{(-+-)}\frac{n^--n^+}{(\Delta-\omega_1-\omega_2)(\Delta-\omega_1)}+S^{kij}_{(+-+)}\frac{n^+-n^-}{(\Delta+\omega_1+\omega_2)(\Delta+\omega_1)}$$
$$+S^{ijk}_{(-+-)}\frac{n^+-n^-}{(\omega_1+\Delta)(\omega_2-\Delta)}+S^{jki}_{(-+-)}\frac{n^--n^+}{(\Delta+\omega_1+\omega_2)(\Delta+\omega_2)}\Big\}, \tag{19}$$

where $n^{\pm}=n_F[E_{(\pm)}(\mathbf{p})]$, $\Delta=E_{(+)}(\mathbf{p})-E_{(-)}(\mathbf{p})$. The expression for diagram *d* is obtained from this by the interchange $\omega_1 j\leftrightarrow\omega_2 k$.

We assume the spin-orbit energy to be small compared both with the Fermi energy and with the radiation-photon energy:

$$\alpha p_F/\hbar \ll \varepsilon_F,\ \hbar\omega. \tag{20}$$

In this case the sum of the triangular diagrams reduces at $\omega_1=\omega_2=\omega$ to the expression

$$\frac{\alpha^3}{\pi^2\omega^2}\int d^2p(n^--n^+)\Big\{[\mathbf{cP}]^i(\delta^{jk}-[\mathbf{cP}]^j[\mathbf{cP}]^k)$$
$$+\frac{1}{2}[\mathbf{cP}]^j(\delta^{ki}-[\mathbf{cP}]^k[\mathbf{cP}]^i)+\frac{1}{2}[\mathbf{cP}]^k(\delta^{ij}-[\mathbf{cP}]^i[\mathbf{cP}]^j)\Big\}. \tag{21}$$

It is seen from Eqs. (14) and (15) that the value of this integral depends on the ratio of the spin-orbit energy $\alpha p_F/\hbar$ to the paramagnetic splitting energy $\hbar\omega_P=2gH$. We consider only the case $\hbar\omega_P<\alpha p_F/\hbar$, which admits of arbitrarily weak fields. Evaluating the integrals in (21), we obtain

$$\mathbf{J}_l^{(2)} = -\frac{2m}{\pi}\left(\frac{e\alpha}{\hbar^2\omega}\right)^3\frac{g}{\hbar\omega}[\mathbf{cH}]\mathbf{E}^2. \tag{22}$$

The expression for the current might contain, in principle, one more vector combination $\mathbf{E}(\mathbf{E}\cdot\mathbf{c}\times\mathbf{H})$. In principal order in $\alpha p_F/\hbar^2\omega$, however, the coefficient of this combination is zero. The fact that the current can contain only odd powers of the spin-orbit interaction constant α can be understood for all calculations. The Hamiltonian and the current operator do not go over into one another under the inversion transformation P: $\mathbf{r}\to-\mathbf{r}$. If, however, we introduce a generalized transformation P that changes in addition the sign of the vector $\mathbf{c}$ (or, equivalently, the sign of the constant α), the Hamiltonian becomes even in $\tilde{P}$, and the current opperator odd. It follows hence that the expectation value of the current contains no even powers of α.

It can be seen from (10) that spin fluxes are capable of contributing to an electric current via spin-orbit interaction. Moreover, in the case $\omega\tau\gg1$, the entire linear current is due only to these fluxes. The physical cause of its onset is that, notwithstanding the translational invariance of the Hamiltonian (8), the total-current operator does not commute with the Hamiltonian. This means that the quadratic fluctuation of the current in the ground state does not vanish, in analogy with the situation in quantum electrodynamics. The nonlinear current is produced as a response of these fluctuations to an external magetic field and to the electromagnetic-wave field.

4. CONCLUSION

If the incidence of the light wave on the layer is not strictly normal, the usual nonlocal current differs from zero[1,8]:

$$\mathbf{J}_{nl}^{(2)} = -\frac{e^3n}{2m^2\omega^3}\mathbf{q}_{\|}\mathbf{E}^2, \tag{23}$$

where n is the electron density and $\mathbf{q}_{\|}$ is the component of the wave vector $\mathbf{q}$ along the plane. In this case, generally speaking, the current component perpendicular to the layer also differs from zero.

The nonlocal current is much larger than the local one for practically all incidence angles. The ratio of these currents can be reduced to the form

$$\frac{J_l^{(2)}}{J_{nl}^{(2)}} = \frac{gH}{2\pi\hbar\omega}\left[\left(\frac{\alpha p_F/\hbar}{\varepsilon_F}\right)^3\left(\frac{p_F}{\hbar}\right)^3\frac{c}{n\omega}\right]\left(\frac{q}{q_{\|}}\right), \tag{24}$$

where c is the speed of light. To increase this ratio it is helpful to use low frequencies ω. We have neglected everywhere in the foregoing the influence of the impurities, for which purpose the condition $\omega\tau\gg1$ must hold. Although there is as yet no complete theory with allowance for impurities, there are grounds for assuming that their influence will lead to an increase of the local current.

In a gallium-arsenide structure with electron mobility $\mu=10^6$ cm^{-2}M/V·s, density $n=0.4\cdot10^{12}$ cm^{-2}, and $\alpha=2.5\cdot10^{-10}$ eV·cm (Ref. 4), at an infrared frequency $\hbar\omega=10$ meV and a magnetic field $H=20$ kOe, we have $\hbar/\tau=0.14$ meV, $\alpha p_F/\hbar=0.4$ meV, and $gH/\hbar\omega=1.2\cdot10^{-3}$, and the factor in the square brackets of (24) is equal to 0.8. We obtain thus $J_l^{(2)}/J_{nl}^{(2)}=1.5\cdot10^{-4}\ (q/q_{\|})$. Assume that the incidence can be made normal accurate to 10^{-3}, and then $J_l^2/J_{nl}^{(2)}\approx0.1$. We take next into account the following important circumstance: the currents $J_l^{(2)}$ and $J_{nl}^{(2)}$ have different directions in the plane, so that they emissions are differently polarized. Thus, by choosing the reflected-light observation direction, for example, to be likewise almost normal to the surface and passing the reflected light through a polarizer, we can decrease the influence of $J_{nl}^{(2)}$ even more.

In addition, the square of the total current, and hence the intensity of the reflected radiation, contain as a term the product

$$\mathbf{J}_l^{(2)}\mathbf{J}_{nl}^{(2)} \sim \mathbf{q}_{\parallel}[\mathbf{cH}]. \tag{25}$$

This addition is odd in the magnetic field direction, the layer orientation, and the wave vector. The appearance of this dependence is typical of systems without inversion centers in a magnetic field. Thus, a correction of this type to the energy of an elementary excitation was investigated in Refs. 9 and 10 for excitons in CdS crystals and in Ref. 11 for light scattering by free carriers in CdS with spin flip. A similar dependence of the intensity of the spin resonance on electrons was observed in InSb (Ref. 12).

Notice must be taken of the close analogy between the considered frequency-doubling phenomenon and the process of light-stimulated direct current (the photovoltaic effect[13]), which can also occur in the described situation.

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