Multiphonon cascade process and secondary radiation bands in polar semiconductors

I. G. Lang, S. T. Pavlov, and G. Yu. Yashin

A. F. Ioffe Physicotechnical Institute, USSR Academy of Sciences
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A new type of resonant Raman scattering of light in a semiconductor exposed to light of frequency $\omega_\lambda > E_g/\hbar + (1+m_e/m_h)\,\omega_{LO}$ is predicted. The secondary radiation occupies the frequency band $0<\omega_s<(1+m_e/m_h)^{-1}(\omega_l-\hbar^{-1}E_g)-\omega_{LO}$. It is shown that the cross section has a steplike dependence on both ω_s and ω_l.

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In this paper we predict theoretically a new type of secondary radiation of semiconductors. The process can be represented as a succession of the following real transitions: primary radiation of frequency ω_l produces an electron-hole pair (EHP); the electron (or hole) emits in succession several LO phonons; finally, the electron (or hole) emits a secondary-radiation quantum accompanied also by one optical phonon. The last step–indirect emission of light by the electron–can be regarded as the inverse of indirect absorption of light by free carriers that interact with LO phonons.[1] A cascade of successive real transitions, wherein the electron emits LO phonons, was theoretically considered in[2] (see also[3,4]) with an aim at a description of multiphonon resonant Raman scattering (MPRRS) of light, i.e., scattering with frequency $\omega_l-n\omega_{\rm LO}$, where n is an integer. The process described here differs from MPRRS in that the radiation in the last stage is produced without annihilation of an EHP. The frequency ω_s of the secondary radiation is, of course, much lower than the frequency $\omega_l-n\omega_{\rm LO}$ and occupies an entire frequency band, because the kinetic energy of the electron or hole in the final state can be different.

We consider a straight-band polar semiconductor in which $m_e<m_h$, where $m_e(m_h)$ is the electron (hole) effective mass. If such a semiconductor is exposed at a temperature much lower than the Debye temperatures to light of frequency

$$\omega_l > E_g/\hbar + (1 + m_e/m_h)\omega_{LO}, \tag{1}$$

where E_g is the width of the forbidden band, secondary radiation is produced in a band

$$0 < \omega_s < (1 + m_e/m_h)^{-1}(\omega_L - E_g/\hbar) - \omega_{LO}. \tag{2}$$

For the sake of simplicity we disregard the dispersion of the optical phonons.

The cross section, averaged over the polarizations of the incident light and summed over the polarizations of the secondary radiation, of a process in which an electron produces together with the secondary radiation only one LO phonon, is defined by the expression

$$\frac{d^2\sigma_1}{d\Omega\, d\omega_s} = \frac{V_0^2\,\omega_s^2}{(2\pi)^3 c^4}\, W_p(\omega_l)\, \frac{W_1\left(E_e, \frac{E'_e}{E_e}, \frac{2}{3}\right)}{W_\tau(E_e)}, \tag{3}$$

where V_0 is the normalization volume, c is the speed of light, $E_e=(1+m_e/m_h)^{-1}\times(\hbar\omega_l-E_g)$ is the kinetic energy of the electron produced by the primary radiation, $E'_e=E_e-\hbar\omega_{\rm LO}-\hbar\omega_s$ is the kinetic energy of the electron in the final state, $W_p(\omega_l)$ is the number of EHP produced per unit

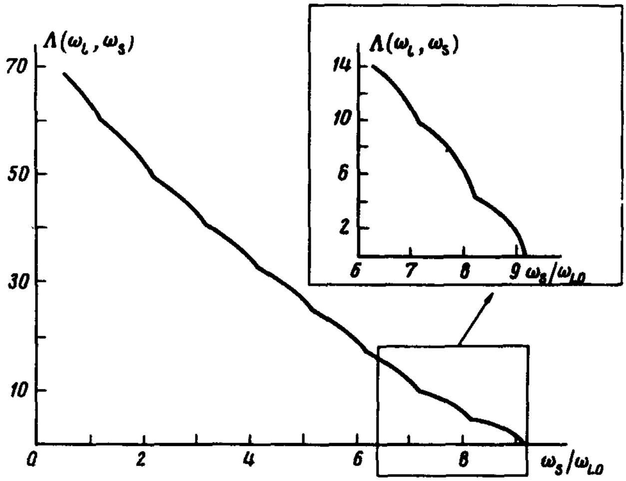

FIG. 1. Dependence of the dimensionless quantity

$$\Lambda(\omega_l, \omega_s) = \left(\frac{e^2 \omega_{LO} V_0 k(\omega_l)}{8\pi^2 m_e c^3 \omega_s n(\omega_l)}\right)^{-1} \frac{d^2\sigma}{d\Omega d\omega_s} \text{ on } \omega_s/\omega_{LO} \text{ at } m_e/m_h = 0{,}2;$$
$$\hbar\omega_l - E_g = 12.4\, \hbar\omega_{LO}$$

time if the volume V_0 contains one photon with energy $\hbar\omega_l$, $W_1(E_e, E'_e/E_e, \sin^2\eta)$ is the probability that an electron with energy E_0 and momentum $\mathbf{p}$ emits in a unit time one photon with wave vector $\vec{\kappa}_s$ and energy $\hbar\omega_s$ and one LO photon, η is the angle between the vectors $\mathbf{p}$ and $\vec{\kappa}_s$, and $W_\tau(E_e)$ is the total reciprocal lifetime of an electron with energy E_e. In lowest-order perturbation theory we have

$$W_1(E_e, E'_e/E_e, \sin^2\eta) = \frac{2\pi e^2 \omega_{LO}^{3/2} \alpha_e}{V_0 m_e \omega_s^3 \hbar^{1/2}} E_e^{1/2} f\left(\frac{E'_e}{E_e}, \sin^2\eta\right), \tag{4}$$

where e is the electron charge, α_e is the Froehlich electron-phonon coupling constant, and

$$f(x, \sin^2\eta) = x^{1/2}(1+x) - \frac{1}{2}(1-x)\ln\frac{1+x^{1/2}}{1-x^{1/2}} + \left[\frac{1}{2}x^{1/2}(5-3x) + \frac{3}{4}(1-x)^2 x \ln\frac{1+x^{1/2}}{1-x^{1/2}}\right]\sin^2\eta. \tag{5}$$

If the interaction of the electron with the LO phonon is the strongest one, then $W_\tau(E_e)$ is determined by the LO-phonon emission probability, and is equal to[5]

$$W_\tau(E_e) = 2\alpha_e \omega_{LO}\left(\frac{\hbar\omega_{LO}}{E_e}\right)^{1/2} \mathrm{Arch}\left(\frac{E_e}{\hbar\omega_{LO}}\right)^{1/2} \tag{6}$$

We note that when (5) and (6) are substituted in (3), the cross section (3) does not depend on the

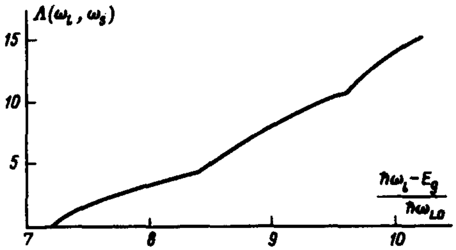

FIG. 2. Dependence of the dimensionless quantity

$$\Lambda(\omega_l, \omega_s) = \left(\frac{e^2\omega_{LO}V_0\, k(\omega_l)}{8\pi^2 m_e c^3 \omega_s\, n(\omega_l)}\right)^{-1} \frac{d^2\sigma}{d\Omega d\omega_s}$$

on $(\omega_l - E_g/h)/\omega_{LO}$ at $m_e/m_h = 0{,}2$; $\omega_s = 6\omega_{LO}$

value of the constant α_e. For quantitative estimates it is convenient to use the expression

$$W_p(\omega_l) = \frac{k(\omega_l)}{n(\omega_l)}c, \tag{7}$$

where $k(\omega_l)$ is the coefficient of light absorption on account of direct EHP production and $n(\omega_l)$ is the refractive index.

At

$$\omega_l > E_g/\hbar + n(1 + m_e/m_h)\omega_{LO}, \tag{8}$$

where $n > 1$, more complicated processes are possible: the electrons produced by light of frequency ω_l can emit in succession $k < n$ LO phonons, and only then can it emit light simultaneously with one more LO phonon. The contribution of these processes to the cross section are not small in comparison with the "single-phonon" cross section (3). To calculate the cross section it is convenient to use the initial formula and the diagram technique proposed in[6,7]; in the simplest cases the same results can be obtained from the balance equations.

The total cross section, which describes processes in which the electrons and holes emit any number of phonons admissible by the energy conservation law, is given by

$$\frac{d^2\sigma}{d\Omega d\omega_s} = \frac{e^2\omega_{LO}V_0\, k(\omega_l)}{8\pi^2 m_e c^3\omega_s\, n(\omega_l)}\left\{\sum_{k<E_e/\hbar\omega_{LO}-1}\left(\frac{E_e}{\hbar\omega_{LO}} - k\right)\right.$$

$$\times f\left(1 - \frac{1+\omega_s/\omega_{LO}}{E_e/\hbar\omega_{LO} - k};\ \frac{2}{3}\right)\left[\mathrm{Arch}\left(\frac{E_e}{\hbar\omega_{LO}} - k\right)^{1/2}\right]^{-1}$$

$$+\frac{m_e}{m_h}\sum_{k<E_h/\hbar\omega_{LO}-1}\left(\frac{E_h}{\hbar\omega_{LO}} - k\right)f\left(1 - \frac{1+\omega_s/\omega_{LO}}{E_h/\hbar\omega_{LO} - k};\ \frac{2}{3}\right)$$

$$\left.\times\left[\mathrm{Arch}\left(\frac{E_h}{\hbar\omega_{LO}} - k\right)^{1/2}\right]^{-1}\right\}, \tag{9}$$

where $E_h = (1 + m_h/m_e)^{-1}(\hbar\omega_l - E_g)$ is the kinetic energy of the hole produced by light of frequency ω_l. The summation over k in (9) begins with $k = 0$, so that the total number of emitted phonons is $k+1$.

Figure 1 shows the quantity $\omega_s d^2\sigma/d\Omega\, d\omega_s$ calculated with the aid of (9) as a function of ω_s/ω_{LO}. It is seen from Fig. 1 that this dependence has a steplike character. Each higher step corresponds to turning on of a process in which the number of emitted phonons increases by unity. The thresholds correspond to the values

$$\omega_s = \left(1 + \frac{m_e}{m_h}\right)^{-1}\left(\omega_l - \frac{E_g}{\hbar}\right) - (k+1)\,\omega_{LO} \tag{10}$$

Figure 2 shows the dependence of $\omega_s(d^2\sigma/d\Omega\, d\omega_s)n(\omega_l)/k(\omega_l)$ on ω_l/ω_{LO} at a fixed frequency ω_s. This dependence is also steplike, but the distance between the threshold points is different than in Fig. 1, since the thresholds are observed at

$$\omega_l = \frac{E_g}{\hbar} + \left(1 + \frac{m_e}{m_h}\right)\left(1 + k + \frac{\omega_s}{\omega_{LO}}\right)\omega_{LO}. \tag{11}$$

Calculations show that the steps are more strongly pronounced at a small maximally possible number of emitted phonons. At $m_e \ll m_h$ the contribution of the holes to the total cross section is small. Observation of long-wave secondary radiation in different semiconductors would make it possible to determine certain parameters, for example the frequency ω_{LO} and the ratio m_e/m_h [see (10) and (11)]

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