Nonlinear polarization interaction of electrons with short-wave phonons

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A pair of short-wave phonons with small total momentum generates an electric macrofield. The electron-phonon interaction due to this field turns out to be quite appreciable.

The latest experiments on pinning and Raman scattering (see, e.g., [1-3]), in which a large contribution of two-phonon processes is observed, have created interest in nonlinear electron-phonon interaction. The magnitude of this interaction has for a long time been regarded as negligible, once definite mechanisms for its cancellation have been established.[4-6] The giant nonlinear deformation potentials ($D_2 \sim 10^5 - 10^6$ eV), recently introduced[7] to explain the results of[1], were obtained without taking into account Herring's warnings[6] and have already been experimentally refuted.[3]

We consider below a new mechanism of nonlinear interaction, mainly nonlinear electron-phonon polarization interaction, and show that in many cases it should turn out to be dominant.

The point is that an important role is usually played in semiconductors by processes with small transfer of the electron momentum, $|\Delta_{\mathbf{p}}| \ll p_B$, where p_B is the Brillouin momentum. As applied to two-phonon processes, this means that the total transfer of the phonon momentum is $|\mathbf{q}_1 \pm \mathbf{q}_2| \ll p_B$, although the momenta $\mathbf{q}_1$ and $\mathbf{q}_2$ of the individual phonons can be arbitrary in this case (usually $q_1, q_2 \sim p_B$). Under these conditions, each of the phonons can generate only a microfield with a period $q_1^{-1} \sim q_2^{-1} \sim a$ (a is the lattice constant), but a long-wave combination of two phonons should generate a nonlinear dielectric polarization in a macrofield with a period $|\mathbf{q}_1 + \mathbf{q}_2|^{-1} \gg a$. The longitudinal component of this macrofield should strongly interact with electrons. We have previously considered[8] a situation that is to a certain degree related, namely, two TO phonons generate a longitudinal macrofield although each of them individually carries only a transverse field.

A direct indication of the presence of a nonlinear macrofield is the experimentally established activity of different combinations of short-wave phonons with $\mathbf{q}_1$

$+\mathbf{q}_2\approx 0$ in infrared absorption.[9,10] Of course, this activity proved directly only the presence of a transverse macrofield. In a cubic crystal, however, all three components of the macroscopic dipole moment are transformed in accordance with a single representation; therefore, the existence of a transverse macrofield for a definite phonon combination implies also the existence of a longitudinal one.

We present order-of-magnitude estimates. The charge induced in the atom by a shift $\mathbf{u}_1$ corresponding to the phonon $\mathbf{q}_1$ is of the order eu_1/a; therefore the atomic dipole moment generated by the shift $\mathbf{u}_2$ corresponding to $\mathbf{q}_2$ is of the order of eu_1u_2/a. As the result, the dipole-moment density is $P\sim eu_1u_2/a^4$, the scalar potential is $\phi\sim P/q$ ($\mathbf{q}=\mathbf{q}_1+\mathbf{q}_2$), and the Hamiltonian of the nonlinear polarization interaction is expressed symbolically in the form

$$H_2^P=e\phi\sim\frac{e^2}{a^4}\frac{u^2}{q}\sim\frac{E_0}{aq}\left(\frac{u}{a}\right)^2,\quad E_0\sim\frac{e^2}{a}\sim 10\text{ eV}. \tag{1}$$

The Hamiltonians of the nonlinear deformation interaction and of the linear interactions of both types are represented in the form

$$H_2^D\sim E_0\left(\frac{u}{a}\right)^2,\quad H_1^P\sim\frac{E_0}{aq}\frac{u}{a},\quad H_1^D\sim E_0\frac{u}{a}. \tag{2}$$

It is seen from (1) and (2) that $H_2^P/H_2^D\sim(aq)^{-1}\gg 1$ in all cases when transitions with small q predominate.

The effectiveness of different interactions can be estimated from the electron-scattering probability w. In order for the final formulas to contain fewer parameters, it is convenient to assume that the thermal energy and the energies of the electron and phonon are of the same order, $kT\sim\hbar\omega_0\sim p^2/2m$. It then follows from (1) and (2) that

$$w_1^P\sim\mu^{-1}\omega_0,\quad w_1^D\sim w_2^P\sim\mu\omega_0,\quad w_2^D\sim\mu^3\omega_0, \tag{3}$$

where the adiabatic parameter is $\mu\sim(m/M)^{1/4}\sim(\hbar\omega_0/E_0)^{1/2}$. We see that the nonlinear polarization interaction predominates over the nonlinear deformation interaction and is comparable with the linear deformation interaction. Of course, such estimates should not be taken too literally. This is seen from the fact that w_1^P contains in fact, in place of μ^{-1}, the Fröhlich coupling constant α, and usually $\alpha\lesssim 1$. We therefore represent H_2^P below in terms of quantities that can be estimated directly, using data on two-phonon lattice absorption.

The nonlinear lattice polarization is represented in the form

$$\mathbf{P}(\mathbf{r})=\frac{1}{2V}\sum_{\mathbf{q}_1\mathbf{q}_2\sigma\tau} g_{\sigma\tau}(\mathbf{q}_1,\mathbf{q}_2)B_\sigma(\mathbf{q}_1)B_\tau(\mathbf{q}_2)e^{i\mathbf{q}\mathbf{r}},$$
$$B_\sigma(\mathbf{q}_1)=b_\sigma(\mathbf{q}_1)\quad b_\sigma(-\mathbf{q}_1); \tag{4}$$

σ and τ number the branches of the phonon spectrum, V is the normalization volume, and $\mathbf{q}=\mathbf{q}_1+\mathbf{q}_2$. According to the estimate (1), the coefficients are given by $g\sim\hbar\omega_0\alpha^2/e$. At $q\ll p_B$ we have $g_{\sigma\tau}(\mathbf{q}_1\cdot\mathbf{q}_2)=g_{\sigma\tau}(\mathbf{q}_1)$.

A standard calculation, as applied to cubic crystals, yields now the following formula for two-phonon absorption:

$$\epsilon''(\omega)=\frac{1}{6\pi\hbar}G^2(\omega)\rho(\omega), \tag{5}$$

where $\rho(\omega)$ is the density of states

$$\rho(\omega)=\sum_{\sigma\tau}\int d^3\mathbf{q}_1\delta(\omega-\omega_\sigma(\mathbf{q}_1)-\omega_\tau(-\mathbf{q}_1)), \tag{6}$$

and $G^2(\omega)$ is the average of $|g_{\sigma\tau}(\mathbf{q}_1)|^2$ over the equal-energy surfaces $\omega_\sigma(\mathbf{q}_1)+\omega_\tau(-\mathbf{q}_1)=\omega$.

On the other hand, by calculating the scalar potentials produced by the polarization $\mathbf{P}(\mathbf{r})$, we obtain

$$H_2^P=\frac{2\pi e}{iV}\sum_{\mathbf{q}_1\mathbf{q}_2\sigma\tau}\frac{(\mathbf{q}\mathbf{g}_{\sigma\tau}(\mathbf{q}_1))}{q^2}B_\sigma(\mathbf{q}_1)B_\tau(\mathbf{q}_2)e^{i\mathbf{q}\mathbf{r}}. \tag{7}$$

For comparison, it is convenient to introduce the nonlinear deformation potential $D_{\sigma\tau}(\mathbf{q}_1\cdot\mathbf{q}_2)$ in accordance with

$$H_2^D=\frac{u_0^2a}{2V}\sum_{\mathbf{q}_1\mathbf{q}_2\sigma\tau}D_{\sigma\tau}(\mathbf{q}_1\mathbf{q}_2)B_\sigma(\mathbf{q}_1)B_\tau(\mathbf{q}_2)e^{i\mathbf{q}\mathbf{r}},\quad u_0^2=\hbar/M\omega_0, \tag{8}$$

where ω_0 is the characteristic phonon frequency, a^3 is the volume of the cell, and M is the atomic mass. It follows from (7) and (8) that the ratio of the deformation and polarization interaction is determined by comparing $D_{\sigma\tau}$ with

$$D^*_{\sigma\tau}(\mathbf{q}_1\mathbf{q}_2)=\frac{4\pi e}{iu_0^2a}\frac{1}{q^2}(\mathbf{q}\mathbf{g}_{\sigma\tau}(\mathbf{q}_1)). \tag{9}$$

Using the experimental data on two-phonon absorption in Ge[9] near 30 μ and formulas (5), (6), and (9), we easily obtain the estimate $qaD^*(q)\sim 10^3$ eV.

In Raman scattering, an important role is played by virtual transitions with production of electron-hole pairs or excitons. Owing to their electroneutrality, the principal terms proportional to q^{-1} cancel out, which is equivalent to replacement of qa by a quantity on the order of unity.[11] The resultant $D^*\sim 10^3$ eV is close to the values of the deformation-potential constants obtained experimentally in[3].

We note in conclusion that linear electron-phonon interaction, taken in second-order perturbation theory, does not lead to singularities as $q\to 0$, and therefore for nonlinear polarization interaction nothing alalogous to the Holstein compensation takes place.[4]

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ERRATA

Erratum: Nonlinear polarization interaction of electron with short-wave phonons [JETP Lett. 20, 27 (1974)]

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On p. 28 the second part of formula (4) should read:

$$B_\sigma(q_1) = b_\sigma(q_1) + b_\sigma^+(-q_1).$$