the available information on the optical and other properties of defects in Si and GaAs are insufficient for drawing reliable conclusions on the avalanche mechanism; the many-photon ionization has not yet been considered in the literature at all but the sensitivity of the avalanche and many-photon ionization processes to the structure of a crystal is highly likely.

Thus, our experimental results and theoretical estimates suggest that the most probable laser damage mechanism in the case of pure Si and GaAs crystals subjected to CO_2 laser radiation is the avalanche impact ionization. In the case of the damage caused by the radiation of the Er^{3+} lasers, it is more likely to be due to the three-photon ionization process. However, final conclusions on the dominant damage mechanism cannot be drawn without further experiments, especially on the kinetics of excitation of nonequilibrium carriers, and without detailed development of the theory for specific materials.

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Optical characteristics of bound polaron and phonon states

V. I. Mel'nikov

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences

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The change of the phonon spectrum in the presence of a polaron is investigated and the frequencies of the lowest bound states are calculated. The oscillator strengths are estimated for absorption and Raman scattering of light with excitation of bound states of phonons.

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The author and Rashba[1] were the first to consider the restructuring of the phonon spectrum in the presence of a continual tight-binding polaron. It was proved that the change of the polarizability of the crystal near the polaron gives rise to an infinite number of phonon states whose frequencies lie lower than the frequency of the free phonon and differ from it by universal numerical factors. To our knowledge, this was the first example of formation of a bound state of an electron and phonon. It turned out subsequently that such states arise in a large class of cases (see the review[2]).

In all the investigated situations it is possible to explain only the qualitative features of the energy spectrum. The only exception is the autolocalized electron state produced in a contact interaction with phonons in a one-dimensional system,[3] when the problem of the spectrum and wave functions of the bound state has an exact analytic solution.[4] In all other cases it is necessary to resort to numerical methods to obtain quantitative results.

The present paper is a continuation of the earlier[1] investigation of the phonon spectrum in the presence of a polaron, describes the method and results of a numerical calculation of the lowest bound states, and presents an estimate of the oscillator strengths for optical transitions with excitation of these states.

The Hamiltonian of an electron interacting with dispersionless polarization phonons and with an electric field E is of the form

$$H=\frac{p^2}{2}+H_{\text{ph}}^0+(2^{3/2}\pi\alpha)^{1/2}\sum_{\mathbf{k}}\frac{e^{i\mathbf{kr}}}{k}q_{\mathbf{k}}+\frac{e}{m^{1/2}\omega^{3/2}}\mathbf{Er}, \qquad (1)$$

where α is the electron-phonon interaction constant, $\mathbf{p}$ is the electron momentum, q_k is the coordinate of a phonon with momentum $\mathbf{k}$, H_{ph}^0 is the Hamiltonian of the free phonons, the energy is measured in units of ω_0, and the length is measured in units of $(m\omega_0)^{-1/2}$ (ω_0 is the phonon energy and m is the electron mass).

In the strong-interaction limit we can obtain from (1) the Hamiltonian of a polaron interacting with phonons.[5] We repeat here briefly this derivation, with an aim of finding the connection between the electric field and the coordinates of the bound states. A shift of the origin of

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the phonon coordinates, $q_k \to q_k + q_k^0$, leads to the appearance of a potential well made up of the displacements q_k^0. The Hamiltonian (1) can be rewritten in the representation of the wave functions of an electron interacting with this well:

$$H_{nn'} = E_n\delta_{nn'} + H_\phi{}^0\delta_{nn'} + (2^{5/2}\pi\alpha)^{1/2}\sum_{\mathbf{k}}\frac{q_{\mathbf{k}}}{k}(\psi_n e^{i\mathbf{k}\mathbf{r}}\psi_{n'})(1-\delta_{n0}\delta_{n'0}) + \frac{e}{m^{1/2}\omega_0{}^{3/2}}\mathbf{E}(\psi_n\mathbf{r}\psi_{n'}). \tag{2}$$

The displacements q_k^0 are connected with the electron ground-state function by the self-consistency condition[6]

$$q_{\mathbf{k}}{}^0 = -(2^{5/2}\pi\alpha)^{1/2}k^{-1}(\psi_0 e^{i\mathbf{k}\mathbf{r}}\psi_0), \tag{3}$$

and the functions ψ_n satisfy the equation

$$\left(-\frac{\Delta}{2} - \alpha 2^{1/2}\int\frac{d\mathbf{r}'\psi_0{}^2(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\right)\psi_n(\mathbf{r}) = E_n\psi_n(\mathbf{r}), \tag{4}$$

where we have omitted from the energy E_n a constant term of no importance in our case.

The large ($\sim\alpha^2$) distances between the levels E_0 and E_n make it possible to calculate the last two terms of (2) by perturbation theory near the ground state of the crystal. Leaving out the ground state energy E_0, we obtain the effective Hamiltonian of the phonons in the presence of a polaron and an electric field:

$$H_{\text{ph}} = H_{\text{ph}}{}^0 + 2^{5/2}\pi\alpha\sum_{\mathbf{k}\mathbf{k}'}\frac{q_{\mathbf{k}}q_{\mathbf{k}'}}{kk'}\sum_{n>0}\frac{(\psi_0 e^{i\mathbf{k}\mathbf{r}}\psi_n)(\psi_n e^{i\mathbf{k}'\mathbf{r}}\psi_0)}{E_0-E_n} + 2\frac{(2^{5/2}\pi\alpha)^{1/2}e\mathbf{E}}{m^{1/2}\omega_0{}^{3/2}}\sum_{\mathbf{k}}\frac{q_{\mathbf{k}}}{k}\sum_{n>0}\frac{(\psi_0 e^{i\mathbf{k}\mathbf{r}}\psi_n)(\psi_n\mathbf{r}\psi_0)}{E_0-E_n}. \tag{5}$$

The second term (5) corresponds to a change of the polarizability of the crystal in the vicinity of the polaron, and the third to the interaction of light with longitudinal phonons. From (5) we obtain for the wave functions of the bound states the equation

$$\nu^2 q_k = q_k + 2^{7/2}\pi\alpha\sum_{k'}\frac{q_{\mathbf{k}'}}{kk'}\sum_{n>0}\frac{(\psi_0 e^{i\mathbf{k}\mathbf{r}}\psi_n)(\psi_n e^{i\mathbf{k}'\mathbf{r}}\psi_0)}{E_0-E_n}. \tag{6}$$

The successive substitutions

$$q_{\mathbf{k}} = k\varphi(\mathbf{k}),\quad \varphi(\mathbf{r})\psi_0(\mathbf{r}) = \left(-\frac{\nabla^2}{2} - \alpha 2^{1/2}\int\frac{d\mathbf{r}'\psi_0{}^2(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\right)\psi_0 f = -\frac{\psi_0}{2}(\nabla^2 + 2(\nabla\ln\psi_0)\nabla)f \tag{7}$$

make it possible to calculate the infinite sum in (6) and write down the obtained equation in the form of the variational principle[1]

$$1-\nu^2 = \min\left\{16\pi\int\psi_0{}^2(\nabla f)^2 d\mathbf{x} \Big/ \int[\nabla(\nabla^2 + 2(\nabla\ln\psi_0)\nabla)f]^2 d\mathbf{x}\right\}, \tag{8}$$

where we put $\mathbf{r} = \mathbf{x}/\alpha 2^{1/2}$. We note that the right-hand side of (8) does not contain symbolic parameters and is fully defined if the function ψ_0 is known.

Pekar has indicated that the frequencies of the three oscillations corresponding to the shift of the polaron as a whole should vanish.[6] A shift by a distance δR adds a term $i\delta\mathbf{R}\cdot\mathbf{k}q_k^0$ is added to the values of q_k^0 given by (3), so that the functions $k_x q_k^0$, $k_y q_k^0$ and $k_z q_k^0$ are eigenfunctions, corresponding to $\nu = 0$, of Eq. (6). For the corresponding functions f_x, f_y, and f_z we obtain, taking into account the substitutions (7) and the relations obtained after differentiating (4) with respect to x, y, and z, the following simple expressions:

$$f_x,\ f_y,\ f_z = \frac{\partial\ln\psi_0}{\partial x},\ \frac{\partial\ln\psi_0}{\partial y},\ \frac{\partial\ln\psi_0}{\partial z}. \tag{9}$$

We point out a number of relations that must be satisfied by the functions q, φ, and f defined by Eq. (6) with the substitutions (7). Transforming (6) with (7) taken into account, we obtain

$$(1-\nu^2)\nabla^2\varphi = 2^{7/2}\pi\alpha\sum_{n>0}\frac{\psi_0(\mathbf{r})\psi_n(\mathbf{r})(\psi_0\varphi\psi_n)}{E_0-E_n} = -2^{7/2}\pi\alpha\psi_0{}^2 f, \tag{10}$$

from which follows, by virtue of the orthogonality of ψ_0 and ψ_n, the relation

$$\int\psi_0{}^2 f\,d\mathbf{r} = 0, \tag{11}$$

which imposes an additional condition on the functions f with zero angular momentum. This condition is important because the functional (8) contains at $l = 0$ only derivatives of f, and defines by the same token f, apart from a constant that must be determined from (11).

We determine the normalization of the phonon wave function $q(\mathbf{r})$ from the condition that in the ground state the potential energy of the oscillation with frequency ν is equal to $\nu/4$. Taking (6) into account, this condition takes the form

$$\int q^2(\mathbf{r})\,d\mathbf{r} = \frac{1}{2\nu}. \tag{12}$$

The function $q(\mathbf{r})$ is expressed in terms of the function f by the relation

$$q(\mathbf{r}) = \frac{2^{5/2}\alpha}{\pi(1-\nu^2)}\int\frac{\psi_0{}^2(\mathbf{r}')f(\mathbf{r}')\,d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|^2}. \tag{13}$$

At large r, the function $q(\mathbf{r})$ decreases like r^{-l-2} if $l > 0$. At $l = 0$ it is necessary to take (11) into account, so that $q(\mathbf{r}) \propto r^{-4}(l = 0)$.

The essential first step towards the solution of the variational problem (8) is to find the wave function ψ_0 defined by Eq. (4) at $n = 0$. This equation is nonlinear in ψ_0, and was therefore solved by a combination of an iteration method. Namely, we use besides Eq. (4) a variational principle that follows from (4) for the energy E_0. The function ψ_0 was specified in the form

$$\psi_0(x) = \gamma^{3/2}\Psi_0(\gamma x), \tag{14}$$

$$\Psi_0(x) = e^{-x/2}P(x), \tag{15}$$

where $P(x)$ is a power series in x.

From the variational principle for E_0 we obtain the value of the scale factor γ, corresponding to the minimum energy at the given coefficients of the power series (15). The obtained ψ_0 is used to calculate the potential in the parentheses in the left-hand side of (4). The linear equation with this potential is solved by the method of moments, thus determining the coefficients of the power series (15). Using the variational principle again, the scale factor γ is determined and the process continued until the coefficients of the series (15) and the scale factor γ reach stable values.

The described iteration process converges very rapidly for the energy, but the determination of the series coefficients requires several thousand iterations. This is precisely why the number of independent coefficients of the series (15) is limited to three. The function Ψ_0 used in the calculation is of the form

$$\Psi_0(x)=\frac{e^{-x/2}}{(4\pi)^{1/2}}\left[C_1\left(1+\frac{x}{\;}-\frac{x^3}{\;}\right)+C_2\left(x^2+\frac{x^3}{2}\right)+C_3x^4\right]. \tag{16}$$

The values obtained for the coefficients C_i and for γ are

$$C_1=0.1338, \quad C_2=0.01327, \quad C_3\sim10^{-7}, \quad \gamma=1.465. \tag{17}$$

The choice of the function Ψ_0 in the form (16) ensures the absence of a linear and cubic term in its expansion in small x. The importance of this condition can be verified with the only known solution of the variational problem (8). Namely, the right-hand side of (8) should become equal to unity upon substitution of the function $f_x=\partial\ln\psi_0/\partial x$. From the form of (8) it follows that the choice of Ψ_0 in Gaussian form with subsequent substitution of f_x in (8) leads to $\nu=1$ in place of $\nu=0$. The choice of Ψ_0 in a form that ensures the absence of a linear term in the expansion of x with three independent coefficients of the power series leads to a right-hand side of (8) with a value close to 0.4. The reason is that upon substitution of the indicated form of f_x the functional (8) contains derivatives of Ψ_0 up to fourth order, and by the same token turns out to be most sensitive to the behavior of Ψ_0 at short distances. The function Ψ_0 defined by (16) and (17) yields for (8) a right-hand side close to 0.96, i.e., differing by approximately 4% from the exact value.

The described method of estimating the accuracy of Ψ_0 is quite satisfactory if the values of x that are vital in the problem are of the order of unity. With increasing serial number of the state, however, or with increasing angular momentum l, large values of x become important, and this calls for substituting in (8) a function Ψ_0 that is highly accurate at $x\gg1$. The accuracy of Ψ_0 as given by (16) and (17) at $x\gg1$ is estimated in the following manner: At $r\gg\alpha^{-1}$ the potential in (4) turns into a Coulomb potential, so that the difference between the asymptotic form of Ψ_0 and the asymptotic form of the hydrogen function is determined by the difference between the eigenvalue of (4) and the eigenvalue in the hydrogen-atom problem. The asymptotic form obtained for Ψ_0 by the condition that the results can be joined with the solution (16) and (17) is

$$\Psi_0(x)\approx\frac{0.1894}{(4\pi)^{1/2}}e^{-0.389}x^{0.753}(1-0.79/x), \tag{18}$$

and the joining point corresponds to $x=5.23$. The next higher powers of $1/x$ in the parentheses of (18) have small numerical coefficients and can be neglected at $x>5$. Comparison of the numerical solution (16) with the asymptotic form (18) shows that in the interval from $x=5$ to $x=20$ they differ by not more than 7%. The accuracy of the function Ψ_0 obtained by us can therefore be regarded as satisfactory for the determination of the energies of the lowest bound states corresponding to small values of the angular momentum l.

To calculate ν we use the following approximations of the radial parts f' of the functions f:

$$\begin{gathered} f^r=e^{-x/2}\left[C_1\left(1+\frac{x}{2}\right)+\sum_{m=2}^{N}C_mx^m\right], \quad l=0, \\ f^r=e^{-x/2}\sum_{m=0}^{N-1}C_mx^{m+l}, \quad l\geqslant1. \end{gathered} \tag{19)(20}$$

These equations are substituted in the functional (8) and variation with respect to C_m yields a system of linear equations of order N, whose eigenvalues determine the frequencies of the bound states. We give the frequencies of the two lowest states for the first three angular-momentum values:

$$\begin{aligned} &l=0: \quad \nu_1\approx0.80; \quad \nu_2\approx0.97. \\ &l=1: \quad \nu_1=0; \quad \nu_2\approx0.94. \\ &l=2: \quad \nu_1\approx0.93; \quad \nu_2\approx0.97. \end{aligned} \tag{21}$$

We see that the frequencies condense rapidly towards the frequency of the free phonon. These frequency values were obtained at $N=8$. The calculation of the frequencies of the following states and of states with angular momenta $l>2$ is difficult because of the insufficient accuracy of the previously obtained function Ψ_0 when $x\gg1$.

The matrix element of the coordinate for the transition with excitation of a bound state of the phonon is determined by the coefficient of E in the last term of (5), if we substitute for q_k a wave function satisfying Eq. (6) and normalized by relation (12). Taking the subsitution (7) into account, we get

$$\begin{aligned} \mathbf{r}_s&=\frac{2(2^{5/2}\pi\alpha)^{1/2}}{m^{1/2}\omega_0^{3/2}}\sum_{\mathbf{k}}\frac{q_s(\mathbf{k})}{k}\sum_{n>0}\frac{(\psi_0e^{i\mathbf{kr}}\psi_n)(\psi_n\mathbf{r}\psi_0)}{E_0-E_n} \\ &=-\frac{2(2^{5/2}\pi\alpha)^{1/2}}{m^{1/2}\omega_0^{3/2}}\int\psi_0^2\mathbf{r}f_s\,d\mathbf{r}. \end{aligned} \tag{22}$$

It follows therefore that the light excites only states with $l=1$. For the oscillator strength F_s, which determines the intensity of the light absorption when the state f_s is excited, we obtain after changing over to the polaron unit of length ($r\to x/2^{1/2}\alpha$) the expression

$$F_s=\frac{16\pi}{\alpha^4}\left(\int\psi_0\mathbf{x}f_s\,d\mathbf{x}\right)^2\Big/\int[\nabla(\Delta+2(\nabla\ln\psi_0)\nabla)f_s]^2d\mathbf{x}. \tag{23}$$

To separate explicitly the exponent of α, this expression is written in a form that does not depend on the normalization of f_s. It is seen that with increasing binding force the oscillator strength decreases like α^{-4}.

To estimate the intensity of Raman scattering, we use formula (25) of Rashba's paper.[7] According to this formula the imaginary part of the polarization operator, which determines the Raman-scattering intensity per polaron, is given by

$$\operatorname{Im}\Pi_{pol}=\pi\sum_{s}\sum_{e'}\int\frac{d\mathbf{k}'}{2\pi}|F_0(\mathbf{ke}|\mathbf{k}',\mathbf{e}')|^2|q_s(\mathbf{k}-\mathbf{k}')|^2\delta(\omega-\omega'-\omega_s), \tag{24}$$

where F_s is the constant of the interaction of the light with the phonons, and **k** and **e** are the wave vector and polarization vector of the photon. Since the photon wave vectors **k** and **k**′ are very small, it suffices to calculate q_s at $k=0$. This can be easily done by using (6) and (7). The result is

$$q_s(k=0)=\frac{2^{7/2}i\pi\alpha}{1-\nu_s^2}\sum_{\mathbf{k}'}\frac{q_s(\mathbf{k}')}{k'}\sum_{n>0}\frac{(\psi_0 r_x\psi_n)(\psi_n e^{i\mathbf{kr}}\psi_0)}{E_0-E_n}=\frac{(2^{5/2}\pi\alpha)^{1/2}}{1-\nu_s^2}x_s, \tag{25}$$

where x_s is the coordinate matrix element given by (22) for the transition with excitation of the state q_s.

Since $x_s\sim\alpha^{-2}$, the ratio of the Raman-scattering intensity per polaron to the scattering intensity per crystal unit cell is of the order of

$$(\alpha^2 m\omega_0 a^2)^{-3/2}\gg 1, \tag{26}$$

where a^3 is the volume of the unit cell. The inequality sign accounts for the fact that we are working within the framework of the continual model polaron model, for which the applicability criterion is in fact (26). Thus the cross section for Raman scattering with excitation bound states of phonons is gigantic, as is the cross section for the absorption of light.

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Calculation of the energy of the molecular phase of solid hydrogen allowing for three-particle interactions

S. I. Anisimov and Yu. V. Petrov

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences, Moscow

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A calculation is made of the static energy, energy of zero-point vibrations, thermodynamic potential, and equation of state of solid molecular hydrogen in the volume range 10–60 a.u./atom. The calculations are carried out in the group approximation and third-order terms are included. It is shown that, for given values of the specific volume, allowance for the three-particle interactions reduces the energy and pressure compared with the pair approximation. Estimates are given of the precision and limits of validity of earlier pair-approximation calculations [V. P. Trubitsyn, Sov. Phys. Solid State **8**, 688 (1976); V. Magnasco and G. F. Musso, J. Chem. Phys. **47**, 1723 (1967); G. A. Neece, F. J. Rogers, and W. G. Hoover, J. Comput. Phys. **7**, 621 (1971); Hover *et al.* Phys. Earth Planet. Inter. **6**, 60 (1972); England *et al.* Phys. Rev. Lett. **32**, 758 (1974)].

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In calculating the thermodynamic functions of the molecular phase of solid hydrogen, the main task is to determine the static energy of a crystal. This is usually done in the approximation of a pair interaction between the molecules forming the crystal.[1-5] When only the interaction between the nearest neighbors is taken into account in this approximation, the lattice energy per atom, measured from the energy of a free molecule, is given by

$$E=\frac{1}{2n}\sum_{\alpha=1}^{n}\left(\frac{1}{2}\sum_{j\neq\alpha}U_{\alpha,j}^{(2)}\right). \tag{1}$$

Here, n is the number of molecules in a unit cell; the index α labels the molecules in this cell and the index j identifies all the nearest neighbors of this molecule; $U_{\alpha,j}^{(2)}$ is the potential energy of the pair interaction between the molecules.

The expression (1) is the first (binary) term of the expansion of the energy as a series in groups of two, three, etc., molecules. It is shown by Nosanow[6] that the general form of this expansion for the bound-state energy of a system of N particles is

$$E=\sum_{\{k\}\subset N}\mathscr{E}_k(\{k\}). \tag{2}$$

The summation in Eq. (2) is carried out over all the k-particle subgroups of the original set of N particles.

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