Impurity electroabsorption and photodestruction of negative ions in an electric field

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Impurity absorption in a solid and the photodestruction of negative ions in the presence of a uniform electric field are discussed. It is shown that an analytic expression can be obtained for the electroabsorption cross section for a short-range potential and an arbitrary number and disposition of the interaction centers. Electroabsorption by weakly bound *s* and *p* electrons is analyzed in detail, and expressions are given for the subthreshold resonance two-impurity absorption. The results obtained can be used for spectroscopic purposes. The effect of an external electric field on multiphoton ionization is discussed. Analytic expressions are obtained for the dynamic polarizability of an ion in a field and for the probability of multiphoton electroabsorption. It is shown that the number of photons absorbed during ionization in a strong electric field is a nonmonotonic function of the field.

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1. INTRODUCTION

Photoabsorption in external fields is being extensively investigated at present. The high sensitivity of electro- and magneto-optical phenomena to the parameters of free charge carriers in solids is being widely used for spectroscopic purposes. An extensive literature is already available in this field. Analogous processes involving the participation of localized (impurity) states have begun to be investigated both theoretically and experimentally in recent years.

Electroabsorption (EA) by an *s* electron of an impurity center has been discussed by Vinogradov[1,2] in terms of the zero-range potential. Absorption by *p* electrons is also of practical interest. The frequency, field, and polarization characteristics of EA are known to be different for *s* and *p* electrons, and the information provided by the corresponding experiments is also different in scope. These questions are discussed below in Sec. 3. It is also important to note that the overall features of phototransitions in an external electric field, which have so far been investigated for impurity absorption, should also be observed in processes involving the photodestruction of free negative ions. Such experiments do not appear to have been carried out although the high sensitivity of EA to the electron-atom interaction parameters could be used for spectroscopic purposes.

All the Vinogradov results refer to one-impurity EA processes. On the other hand, real crystals practically always contain both deep filled and partially filled levels, and the resonance interaction with the latter may appreciably increase the subthreshold absorption of light in an electric field. This question is discussed below in Sec. 4. The general theoretical description of multi-impurity EA requires the solution of the multicenter problem of absorption of a photon by an electron moving in the resultant potential of fixed scattering centers in the presence of a constant external force. This problem admits of an analytic solution in the case of a short-range interaction (Sec. 2). This approach is generalized below to multiphoton transitions, and this enables us to consider the properties of multiphoton electroabsorption (Sec. 5).

2. SINGLE-PHOTON DESTRUCTION OF NEGATIVE MOLECULAR IONS IN AN ELECTRIC FIELD

The cross section for the photodetachment of an electron in an electric field is given by

$$\sigma=\int d\mathbf{k}\,dE_z\frac{d\sigma}{d\mathbf{k}\,dE_z}\delta\left(E_i+\omega-E_z-\frac{k^2}{2}\right)$$

where $e=\hbar=m=1$. We consider the case of a sufficiently weak field f in which we can neglect the autoionization of the initial state and for which a stationary problem ($f\ll|E_i|^{3/2}$) can be formulated. The differential cross section $d\sigma/d\mathbf{k}dE_z$ refers to an interval of changes in the quantum numbers $\mathbf{k}$ and E_z specifying the state of the electron in the field $\mathbf{f}$ ($\mathbf{f}=0,0,f_z$), $\mathbf{k}=(k_x,k_y,0)$. In the dipole approximation,

$$\begin{gathered}d\sigma/d\mathbf{k}dE_z=4\pi^2\omega|\langle\mathbf{k},E_z|\mathbf{ar}|i\rangle|^2,\\ \langle\mathbf{k},E_z|\mathbf{ar}|i\rangle=\int d\mathbf{r}\,\Psi_{\mathbf{k},E_z}(\mathbf{r})\,\mathbf{ar}\varphi_i(\mathbf{r}).\end{gathered}\tag{1}$$

In these expressions, ω and $\mathbf{a}$ are, respectively, the frequency and polarization vector of the photon, $\mathbf{r}$ is the position vector of the electron, φ_i is the wave function of the initial state, E_i is the corresponding energy ($|E_i|^{3/2}\gg f$), and $\Psi_{\mathbf{k}E_z}$ is the wave function of the liberated electron. In the long-wave approximation, taking into account only *s* scattering of electrons by individual centers (short-range interaction), the wave functions have the following form[3]

$$\varphi_i(\mathbf{r})=N_i\sum_m\operatorname{Re}G^{(+)}(E_i,\mathbf{R}_m,\mathbf{r})\tau_m,\quad\langle\varphi_i|\varphi_i\rangle=1;\tag{2}$$

$$\begin{gathered}\Psi_{\mathbf{k}E_z}(\mathbf{r})=\varphi_{\mathbf{k}E_z}(\mathbf{r})+\sum_m G^{(+)}(E_f,\mathbf{R}_m,\mathbf{r})T_m\varphi_{\mathbf{k}E_z}(\mathbf{R}_m),\\ \varphi_{\mathbf{k}E_z}(\mathbf{r})=N_fe^{i\mathbf{kr}}V[-2(E_z+\mathbf{fr})/(2f)^{2/3}].\end{gathered}\tag{3}$$

In these expressions, $\mathbf{R}_m$ is the position vector of the m-th diffraction center (atom of the molecular or multi-impurity system), $V(Z)$ is the Airy function of the first kind, $E_f = E_z + k^2/2$, $N_{i(f)}$ are normalizing factors, and $N_f^2 = [2\pi^3(2f)^{1/3}]^{-1}$. The functions $\varphi_{\mathbf{k},E_z}(\mathbf{r})$ are normalized so that

$$\langle \varphi_{\mathbf{k},E_z} | \varphi_{\mathbf{k}',E_z'} \rangle = \delta(\mathbf{k}-\mathbf{k}')\,\delta(E_z - E_z'). \tag{4}$$

The function $G^{(+)}(E, \mathbf{r}, \mathbf{r}')$ in (2) and (3) is the three-dimensional Green function for an electron in a uniform electric field:

$$G^{(+)}(E,\mathbf{r},\mathbf{r}') = \int d\mathbf{k}\, dE_z \frac{\varphi_{\mathbf{k},E_z}(\mathbf{r})\overset{*}{\varphi}_{\mathbf{k},E_z}(\mathbf{r}')}{E-E_z-k^2/2+i\eta}$$
$$= \frac{1}{2\pi(2f)^{1/3}|\mathbf{r}-\mathbf{r}'|}\left(\frac{\partial}{\partial\xi}-\frac{\partial}{\partial\zeta}\right)V(Z_1)\,[U(Z_2)+iV(Z_2)], \tag{5}$$
$$\xi,\ \zeta = {}^1/_2(z+z'\pm|\mathbf{r}-\mathbf{r}'|),$$
$$Z_1 = -2(2f)^{-2/3}(E+f\xi), \quad Z_2 = -2(2f)^{-2/3}(E+f\zeta),$$

where $U(Z)$ is the Airy function of the second kind. The quantities τ_m and T_m in (2) and (3) satisfy the set of algebraic equations of the theory of multiple scattering.[3]

For short-range interactions, the coordinate dependence of the wave functions is determined by the Green function. The bilinear combinations of $G^{(+)}(E, \mathbf{R}_m, \mathbf{r})$ in (1), including those containing the electron coordinate (momentum) operator, are most readily integrated with the aid of the spectral representation given by (5), and this enables us to obtain closed analytic expressions for all the quantities characterizing EA. When the energies of the states under consideration are sufficiently low, this approach can be generalized to systems with a resonance interaction (bonding) of p electrons with individual centers. The electron angular momentum can then be taken into account by introducing differential operators that determine the coupling of asymptotic functions for the s and p states [see, for example, Eq. (18) in[4]]. When $l_m = 0$ ($m = 1, \ldots, N$), the normalizing integral of the function given by (2) can be evaluated directly:

$$N_i^{-2} = \sum_{m,m'} \tau_m \tau_{m'} \left(\frac{d}{dE}\operatorname{Re} G^{(+)}(E, \mathbf{R}_m, \mathbf{R}_{m'})\right)_{E=E_i}. \tag{6}$$

The orthogonality of the functions corresponding to the continuous spectrum follows from the equation

$$\langle \Psi_{\mathbf{k},E_z} | \Psi_{\mathbf{k}',E_z'} \rangle = \langle \varphi_{\mathbf{k}E_z} | \varphi_{\mathbf{k}'E_z'} \rangle,$$

which can be established with the aid of equations (4), (5), and (11) in[3]. The wave functions $\varphi_i(\mathbf{r})$ are orthogonal to the functions for the final state $\Psi_{\mathbf{k}E_z}(\mathbf{r})$ to within exponential terms $\sim \Gamma_i$, where Γ_i is the autoionization width of the initial state $\{\Gamma_i \sim \exp[-2(-2E_i)^{3/2}/3f]\}$.

The dipole moment matrix elements for the states (2) and (3), evaluated with the aid of the spectral representation (5) and the orthogonality condition (4), will, in general, have the form

$$\langle \varphi_i | r_\alpha | \Psi_{\mathbf{k},E_z} \rangle = N_i \sum_m \tau_m \left[J_1^\alpha(\mathbf{R}_m) + \sum_{m'} T_{m'} \varphi_{\mathbf{k},E_z}(\mathbf{R}_{m'}) J_2^\alpha(\mathbf{R}_m, \mathbf{R}_{m'}) \right] \tag{7}$$

where $r_\alpha = x, y, z$ for $\alpha = 1, 2, 3$, respectively. Next,

$$J_1^\alpha(\mathbf{R}_m) = -\left[R_m^\alpha + i\frac{k_\alpha}{\omega} \right] \frac{\varphi_{\mathbf{k},E_z}(\mathbf{R}_m)}{\omega}, \tag{8}$$

$$J_2^\alpha(\mathbf{R}_m, \mathbf{R}_{m'}) = \left[R_m^\alpha + \frac{\partial^2}{\partial R_m^\alpha \partial E_f} \right] Q(E_i, E_f, \mathbf{R}_m, \mathbf{R}_{m'}), \quad \alpha = 1, 2, \tag{9}$$

$$Q(E_i, E_f, \mathbf{R}_m, \mathbf{R}_{m'}) = \frac{1}{E_f - E_i} [\operatorname{Re} G^{(+)}(E_i, \mathbf{R}_m, \mathbf{R}_{m'}) - G^{(+)}(E_f, \mathbf{R}_m, \mathbf{R}_{m'})].$$

In these expressions, $\omega = E_f - E_i$ and R_m^α is the α-th projection of the position vector of the m-th center. Finally,

$$J_1^z(\mathbf{R}_m) = \left[\frac{E_z}{f} + \frac{f}{2}\frac{d^2}{dE_z^2} \right] \frac{\varphi_{\mathbf{k}E_z}(\mathbf{R}_m)}{E_z + k^2/2 - E_i}, \tag{10}$$

$$J_2^z(\mathbf{R}_m, \mathbf{R}_{m'}) = \left[R_m^z + \frac{f}{2}\frac{\partial^2}{\partial E_f \partial E_i} \right.$$
$$\left. + \frac{1}{2}\frac{\partial^2}{\partial E_f \partial (R_m^z - R_{m'}^z)} \right] Q(E_i, E_f, \mathbf{R}_m, \mathbf{R}_{m'}). \tag{11}$$

Subsequent integration with respect to $\mathbf{k}$ in (1) can be carried out with the aid of the integral

$$\int x^n f_1(t_1+x) f_2(t_2+x)\, dx = \left(\frac{d^2}{dt_1^2} - t_1\right)^n W(t_1, t_2), \tag{12}$$
$$W(t_1, t_2) = f_1(t_1) f_2'(t_2) - f_1'(t_1) f_2(t_2),$$

where $f_{1,2}$ are any two solutions of the Airy equations. When $t_2 \to t_1$, $f_1 = f_2 = V$, and the formula given by (12) becomes identical with the well-known Aspnes integral[5] (Eq. B24a). For the multicenter short-range interaction model, the above formulas provide the solution for the EA cross section in a general form.

It is important to note, in conclusion, that, whenever the short-range interaction model cannot be entirely justified (high electron binding energy in the initial state), the wave function φ_i can be chosen as a linear combination of Slater orbitals with parameters determined from comparisons with existing variational calculations (this approach to the evaluation of cross sections for the photodestruction of negative atomic ions was used, for example, by Moskvin[6]). All the calculations can be performed by analogy in this case. The final expressions for the matrix elements will contain additional differentiation with respect to the initial-state energy [one can use the obvious result $e^{-\alpha r} = -\partial(e^{-\alpha r}/r)/\partial\alpha$].

3. EA BY A NEGATIVE ATOMIC ION (PHOTODETACHMENT OF s AND p ELECTRONS)

The cross section for the photodetachment of an s electron, calculated from the expressions given in Sec. 2, has the following form for an atomic ion:

$$\sigma(\omega, \mathbf{f}) = \sigma(\omega, 0)\,\Omega(\omega, \mathbf{f}), \tag{13}$$
$$\sigma(\omega, 0) = 4\pi\alpha v^3/3\omega^3, \quad \alpha = (-2E_i)^{1/2},$$
$$v = [2(E_i + \omega)]^{1/2}.$$

In these expressions, σ is the cross section for the photodetection of an s electron in the absence of a field[7] and $\Omega(\omega, \mathbf{f})$ is the EA field factor which depends on the field direction and light polarization.

For unpolarized light and longitudinal field orientation ($\mathbf{a} \perp \mathbf{f}$), we have

$$\Omega_\perp(\omega, \mathrm{f}) = [Z^2V^2 - Z(V')^2 - {}^1/_2 VV']/2Z_0^{1/2}(-Z)^{1/2}V(Z_0)U(Z_0), \tag{14}$$
$$Z_0 = -2E_i/(2f)^{2/3}, \quad Z = -v^2/(2f)^{2/3}$$

where $V=V(Z)$, $V'=V'(Z)$. The expression given by (14) takes into account the distortion of the initial-state wave function by the field. In the lowest-order approximation in the parameter f/α^3, we have $2Z_0^{1/2}V(Z_0)U(Z_0)=1$, and the expression given by (14) becomes identical with the result reported by Vinogradov [Eq. (36) in[1]].

In the case of longitudinally polarized light ($\mathbf{f}\parallel\mathbf{a}$), we have

$$\Omega_{\parallel}(\omega,\mathrm{f})=[2Z_0^{1/2}(-Z)^{1/2}V(Z_0)U(Z_0)]^{-1}\{[Z^2V^2-Z(V')^2-2VV'] +{}^3/_4(2f)^{2/3}V^2\operatorname{Re}L+{}^3/_{16}(2f)^{4/3}|L|^2((V')^2-ZV^2)\}, \tag{15}$$

$$L=\frac{2\pi}{\varkappa-2\pi G_p^{(+)}(E_f)}\left[\frac{d}{dE_f}G_p^{(+)}(E_f)+\frac{d}{dE_i}\operatorname{Re}G_p^{(+)}(E_i)\right].$$

In this expression, $\varkappa^{-1}$ is the scattering length for an s electron on an atom, and

$$G_p^{(+)}(E)=\left[\frac{1}{2\pi|\mathbf{r}-\mathbf{r}'|}-G^{(+)}(E,\mathbf{r},\mathbf{r}')\right]_{r\to r'}$$

is the regularized Green function[1,3,8]

$$2\pi G_p^{(+)}(E)=(2f)^{1/3}\{ZV[U+iV]-V'[U'+iV']\}. \tag{16}$$

It is important to note that the formula given by (15) is not the same as Eq. (37) in[1] (this is probably due to differences in the initial formulation of the problem). In the present paper, we consider a transition from the state $\varphi_i(\mathbf{r})$ localized prior to the interaction with a photon. Vinogradov, on the other hand, considered transitions between stationary states (3) which, strictly speaking, were always unlocalized. This approach corresponds to the free-free transitions of an electron undergoing a resonance interaction with an impurity scattering center.

The role of the electric field is most important in the neighborhood of the threshold frequencies. When $\omega=-E_i$, the turning point of the function given by (3) with respect to the coordinate z is also a particle-localization center in the initial state. Therefore, at the threshold, $\Delta\sigma_\perp=\sigma_\perp(f)-\sigma(0)$ has a maximum (proportional to the first power of the field)

$$\Delta\sigma_\perp(\omega=-E_i)=\sigma_\perp(f)=\frac{4\pi^2\alpha}{9\omega^3}\frac{f}{\Gamma(^1/_3)\Gamma(^2/_3)}. \tag{17}$$

For high ω, the field factors exhibit damped oscillations which, for $f\ll\nu^3$, have the form

$$\begin{aligned}\Omega_\perp&\approx1-\frac{f^2}{8\nu^6}-\frac{3f^2}{4\nu^6}\sin\frac{2\nu^3}{3f},\\ \Omega_\parallel&\approx1+\frac{3f}{2\nu^3}\cos\frac{2\nu^3}{3f}.\end{aligned} \tag{18}$$

Let us now consider absorption by a p electron. Within the framework of the asymptotic theory, all the calculations can be carried out by analogy with the case of an s electron. For the sake of simplicity, we will confine our attention to the derivation of the corresponding formulas for atomic ions. The asymptotic wave function for a state undistorted by the field is

$$\varphi_i^{l=1,\alpha}(\mathbf{r})=N\frac{r_\alpha}{r}\alpha^2K_1(\alpha r)=-N\left[\frac{d}{dR_\alpha}\frac{e^{-\alpha|\mathbf{r}-\mathbf{R}|}}{|\mathbf{r}-\mathbf{R}|}\right]_{\mathbf{R}\to0}, \tag{19}$$

where $\alpha=x,y,z$, and $K_1(\alpha r)$ is the spherical Macdonald function. It is important to note that, in contrast to the $l=0$ case, the normalizing factor in (19) cannot be calculated directly and must be looked upon as an additional phenomenologic parameter.[4] The evaluation of the matrix elements of the momentum operator over the states (19) and (3) can again be carried out with the aid of the spectral representation (5), followed by differentiation with respect to R_α (the limiting transition $\mathbf{R}\to0$ is carried out after differentiation). The final result is

$$\langle\varphi_i^{l=1,\alpha}|r_\alpha|\Psi_{\mathbf{k},E_z}\rangle=\varphi_{\mathbf{k}E_z}(0)\left[\frac{1}{\omega}-\frac{k_\alpha^2}{\omega^2} +t(E_f)\left(1+\frac{\partial^3}{\partial E_i\,\partial R_\alpha^2}\right)Q(E_i,E_f,R_\alpha,0)\right], \tag{20}$$

$$t(E_f)=2\pi[\varkappa-2\pi G_p^{(+)}(E_f)]^{-1}$$

where $\alpha=1,2$. The other matrix elements have the analogous form.

The expression for the total cross section averaged over the components of the electron angular momentum in the initial state is of practical interest. For unpolarized light and longitudinal field orientation ($\mathbf{f}\perp\mathbf{a}$), the average cross section is given by

$$\begin{aligned}\langle\sigma_\perp(f,\omega)\rangle=\frac{2N^2}{3\pi}\frac{(2f)^{1/3}}{\omega^3}&[|\omega+L_1|^2((V')^2-ZV^2)\\ &-{}^2/_3\operatorname{Re}(\omega+L_1)(2f)^{2/3}(Z^2V^2-Z(V')^2-{}^1/_2VV')\\ &+(2f)^{4/3}({}^3/_8V^2-{}^1/_3Z^3V^2+{}^1/_3Z^2(V')^2+{}^1/_6ZVV')],\end{aligned} \tag{21}$$

$$L_1=\frac{2\pi\omega G_p^{(+)}(E_f)-{}^1/_3(\alpha^3-i\nu^3)}{\varkappa-2\pi G_p^{(+)}(E_f)}$$

where $E_i=-\alpha^2/2$ is the energy of a p electron in the initial state. When $f=0$, equation (21) gives the cross section for the photodestruction of an ion with a valence p electron, which is identical with the result obtained by Klein and Brueckner[9] (in the limit as $R=0$, $r_0=0$).

In the neighborhood of threshold frequencies, the quantity $\Delta\langle\sigma_\perp\rangle=\langle\sigma_\perp(f)\rangle-\langle\sigma(0)\rangle$ for a p electron will also have a maximum but, in contrast to the s electron case, the cross section at the threshold is proportional to $f^{1/3}$:

$$\Delta\langle\sigma_\perp(\omega=-E_i)\rangle=\langle\sigma_\perp(f,\omega=-E_i)\rangle=\frac{2N^2}{3\pi\omega}(2f)^{1/3}(V')^2_{(0)}. \tag{22}$$

In this case, the above-threshold oscillations in the cross section for electrophotodestruction depend on the scattering length of the s electron on the atomic core ($\varkappa^{-1}$), and, when $\nu^3\gg f$, $\nu^2\ll\omega$, they are described by

$$\Delta\langle\sigma_\perp(\omega,f)\rangle=C(\omega,\varkappa,E_i)\,f\cos(2\nu^3/3f+\varphi). \tag{23}$$

where

$$\varphi=\operatorname{arctg}\frac{2\varkappa\nu}{\nu^2-\varkappa^2},$$

and $C(\omega,\varkappa,E_i)$ is a slowly-varying function.

Let us now consider the possible use of the above expressions for spectroscopic purposes. The high (phase) sensitivity of the cross sections to the parameters of the electron-atom interaction (such as the electron binding energy and the s scattering length) can probably be used as a basis of a new experimental method of measuring these parameters. This is particularly important in the case of ions with sufficiently high electron binding energy for which the most accurate field ionization method[10] cannot be used in practice. It is important to note that the over-threshold oscillations in (18) and (23) are determined by the interaction of the electron only in the final state, so that the limitations of the short-range potential approximation used in deriving the absolute cross sections are not very important. The spectroscopic possibilities of the electrophotodestruction method are illustrated in Fig. 1 which is concerned with the cross section for the process

$$\mathrm{Au}^- + \omega \to \mathrm{Au} + e^-, \tag{24}$$

calculated from (13) for two tabulated values of the electron binding energy in the Au⁻ ion ([10], p. 64) for a constant field $f = 10^5$ V/cm. Figure 2 illustrates the effect of the external field on the photodestruction cross section, and shows the function $\Delta\sigma_\perp(f) = \sigma_\perp(f) - \sigma(0)$ for different values of f (the experimental values of the photodestruction cross section for the above process are taken from[11]). The difference between the field dependence of EA at near-threshold frequencies [Eqs. (17) and (22)] could be used as a means of determining the configuration (angular momentum) of the states of impurity centers in doped semiconductors.

4. TWO-IMPURITY (RESONANCE) EA

The general formulas given in Sec. 2 can be used to obtain explicit analytic expressions for the total and differential EA cross sections in the case of an arbitrary number and distribution of scattering centers simulating the interaction between an electron and the atomic cores of molecular and multi-impurity systems. We shall use this method to consider two-impurity resonance EA.

The photoeffect does not have a red limit in an electric field. When $\omega < -E_i$, the cross sections for single-impurity absorption are not zero and are given by (13) and (21) with exponentially small field factors. Multi-impurity absorption corresponding to the resonance interaction between the liberated electrons and other impurity atoms become important under these conditions. When an unfilled higher-lying level E_2 is present, the EA cross section will, clearly, have a resonance maximum at the photon frequency $\omega = E_2 + \mathbf{f}\cdot\mathbf{R} - E_1$, where $\mathbf{R}$ is the position vector of the center providing the level E_2, $\mathbf{R}_1 = 0$, and it is assumed that $R \neq 0$. At the resonance frequency, the cross section for the photodetachment of an electron is equal to the cross section for the photoexcitation of a quasilocalized state, i.e., $\sim\exp(-\nu R)$. Thus, the resonance effect becomes appreciable when $f < (E_i + \omega)/R$ $(E_i = E_1)$.

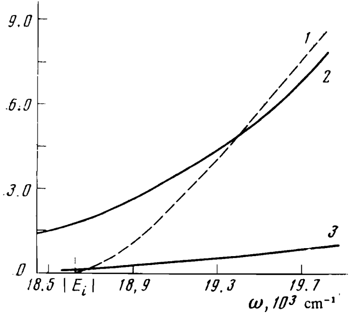

FIG. 1. Photodestruction cross section of the Au⁻ ion in an electric field. 1—Experimental curve for $f = 0$,[11] 2, 3—theoretical curves for $f = 10^5$ V/cm (2—$E_i = -18\,620$ cm⁻¹, 3—$E_i = -22\,650$ cm⁻¹.

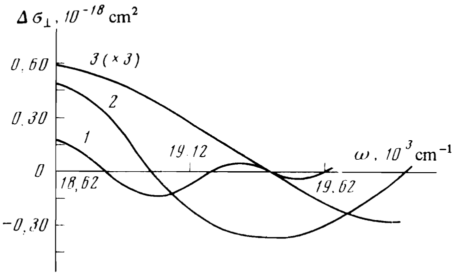

FIG. 2. Dependence of the function $\Delta\sigma_\perp(f, \omega) = \sigma_\perp(f, \omega) - \sigma(0, \omega)$ on photon frequency for different fields f ($E_i = -18\,620$ cm⁻¹): 1—$f = 10^4$ V/cm; 2—$f = 2.8 \times 10^4$ V/cm; 3—$f = 10^5$ V/cm (in the last case, the scale along the ordinate axis is reduced by a factor of three).

The general expression for the total EA cross section in the two-impurity case is rather unwieldy, and we shall therefore confine our attention to a single-resonance term. The cross section for the two-impurity absorption is then

$$\sigma(\omega, R) = \frac{\pi\varkappa_1\varkappa_2\omega}{f} |\Phi(E, R)|^2 \frac{\Gamma_2}{(E_1 + \omega - E_2 - \mathbf{fR})^2 + \Gamma_2^2/4}. \tag{25}$$

In this expression, Γ_2 is the autoionization width of the upper state ($\varkappa_{1,2}^2 = 2|E_{1,2}|$),

$$\begin{gathered} |\Phi(E, \mathbf{R})|^2 = |\Phi_x(E_f, \mathbf{R})|^2 + |\Phi_y(E_f, \mathbf{R})|^2, \\ \Phi_{x,y}(E_f, \mathbf{R}) = \left[R_{x,y} + \frac{\partial^2}{\partial E_f\,\partial R_{x,y}}\right] \frac{2\pi G_p^{(+)}(E_f, \mathbf{R}, 0)}{E_i - E_f} \end{gathered} \tag{26}$$

where $E_f = E_i + \omega$. The above expression describes the resonance two-impurity EA for fixed R (the situation involving an impurity quasimolecule). In the more general case, the EA coefficient is determined by the mean cross section corresponding to the averaging of $\sigma(\omega, R)$ over all the possible relative dispositions of impurities.

In a weak electric field (and neglecting the autoionization of the upper level), the mean cross section is given by

$$\langle\sigma(\omega)\rangle_{\mathbf{R}} = \frac{2\pi^2\varkappa_1\varkappa_2\omega}{f} \int_0^\infty \rho\, d\rho\, |\Phi(\rho, R_z)|^2 n_2(R_z, \rho) \tag{27}$$

where the integral is evaluated over the plane $R_z = (E_1 + E_2 - \omega)/f$. In this expression, n_2 is the concentration of atoms providing the unfilled level E_2. The analytic expression for $\Phi(\rho, R_z)$ is well known [Eq. (26)] and, therefore, Eq. (27) can, at least in principle, be used to determine the function describing the relative distribution of impurities in multicomponent doped semiconductors (such as silicon carbide doped with boron and aluminum[12]).

The asymmetry of the correlation function $n_2(R)$ should have an effect on the EA coefficient as a function of the field direction (when $\mathbf{f}\perp\mathbf{a}$). The mean cross section $\langle\sigma(\omega)\rangle$ regarded as a function of frequency has an asymmetric peak with a maximum at $\omega=E_2-E_1$:

$$\langle\sigma\rangle\approx\frac{n_2\varkappa_1\varkappa_2 f}{8\omega^3}\frac{\exp[-2\nu f^{-1}|E_1+\omega-E_2|]}{|E_1+\omega-E_2|^2}$$
$$\Gamma_2\ll|E_1+\omega-E_2|<|E_1|,\quad n_2\approx\text{const.}$$

The formal singularity at $\omega=E_2-E_1$ is due to the fact that we have neglected autoionization ($\Gamma_2\approx 0$) and the exchange interaction between the states under consideration.

5. MULTIPHOTON ELECTROABSORPTION

Multiphoton EA was first discussed qualitatively by Arutyunyan and Askar'yan.[13] The problem was also considered by Nikishov[14] in the short-range interaction approximation.1) An integral expression was obtained for the multiphoton ionization probability in a uniform electric field [Eq. (23) in Nikishov's paper] but this equation is too complicated for the interpretation of the phenomenon. At the same time, the general conclusion reported in these papers, i.e., that the number of photons absorbed from the electromagnetic wave during ionization should decrease, does not exhaust all the possible field effects. In the analysis given below, we give an analytic expression for the probability of multiphoton EA, which is convenient for qualitative and quantitative interpretations, and show that, in a strong electric field, the effective number n_f of photons absorbed during ionization may increase. This number (degree of nonlinearity of the process) is determined from the dependence of the rate of dissociation of atomic particles on the intensity of the electromagnetic wave.[16]

It is important to note that the matrix element for n-photon EA can be evaluated in an explicit form by using (5) for the function $G^{(+)}$. This approach enables us to obtain an analytic expression for the multiphoton EA probability for an arbitrary orientation of **f**. However, the most interesting case is that of a circularly polarized wave and longitudinal field orientation for which an exact solution is possible because of the symmetry of the interactions (the corresponding solution for $f=0$ was recently obtained by Manakov and Rapoport[17] and by Berson[18]). The energy levels of a weakly bound electron interacting with a circularly polarized wave $\mathbf{F}=F(\mathbf{e}_x\sin\omega t+\mathbf{e}_y\cos\omega t)$ ($\mathbf{e}_{x,y}$ are unit vectors) and an external field **f** are the roots of the equation

$$\varkappa-(-2E)^{1/2}=2\pi G_p^{(+)}(E),\quad \mathbf{fF}=0,\tag{28}$$

where

$$2\pi G_p^{(+)}(E)=\frac{1}{\sqrt{2\pi i}}\int_0^\infty\frac{dt}{t^{3/2}}\exp\left\{i\left(\frac{z^2}{2t}+Et\right)\right\}$$
$$\times\left(1-\exp\left[i\left(\frac{2F^2}{\omega^4 t}\sin^2\frac{\omega t}{2}-\frac{f^2t^3}{24}-\frac{F^2t}{2\omega^2}\right)\right]\right)_{z\to0}.\tag{29}$$

When $f=0$, $F\neq0$, the expression given by (28) is identical with the formula given by (6) in[17]. When $F=0$, $f\neq0$, it becomes identical with the equation given by Demkov and Drukarev.[19]

Let us isolate in the Green function $G_p^{(+)}(E)$ the terms corresponding to a fixed energy of the liberated electron $[E+F^2/2\omega^2+(n-m)\omega]$, and transform it so that it reads as follows:

$$2\pi G_p^{(+)}(E)=-(-2E)^{1/2}+\sum_{n=0}^{\infty}\sum_{m=0}^{2n}C_{2n}{}^m(-1)^m\Omega_{nm}(f,F,\omega),\tag{30}$$

$$\Omega_{nm}(f,F,\omega)=\left(-\frac{2F^2}{\omega^4}\right)^n\frac{1}{n!}\left[\frac{d^n}{da^n}\left(\frac{d}{dZ_1}-\frac{d}{dZ_2}\right)\right.$$
$$\left.\times V(Z_1)(U(Z_2)+iV(Z_2))a^{-1/2}\right]_{a\to0};\tag{31}$$
$$Z_{1,2}=Z_0\pm\frac{(2f)^{1/3}}{2}a^{1/2},\quad Z_0=\frac{-2(E+F^2/2\omega^2+(n-m)\omega)}{(2f)^{2/3}}.$$

Solution of (28) by iteration, in the first order in the terms containing the imaginary part, yields

$$E=E_i-{}^1\!/_2\beta(\omega,f)F^2-i\Gamma/2\quad(E_i=-\varkappa^2/2);\tag{32}$$

$$\beta(\omega,f)=-\frac{1}{\omega^2}-\frac{2\varkappa f}{\omega^4}\sum_{m=0}^{2}C_2{}^m(-1)^m\Phi_{2m}(Z_0),$$
$$\Phi_{2m}(Z_0)=Z_0^2VU-Z_0V'U'-VU'+{}^1\!/_2V'U,\tag{33}$$
$$V=V(Z_0),\quad U=U(Z_0),$$

where β is the dynamic polarizability of the negative ion in a uniform electric field.2) The quantity

$$\Gamma=\frac{\varkappa f}{4\alpha_F^2}\exp\left(-\frac{2}{3}\frac{\alpha_F^3}{f}\right)+\sum_{n=1}^{\infty}\sum_{m=0}^{2n}C_{2n}{}^m(-1)^m\Gamma_{nm}\tag{34}$$

($\alpha_F^2=2E_i+F^2/\omega^2$) is the total ionization width which determines the rate of destruction of the negative ion. The partial widths corresponding to n-photon transitions and the emission of an electron with energy $E=E_i+F^2/2\omega^2+(n-m)\omega$ have the following form:

$$\Gamma_{nm}=\varkappa\,\mathrm{Im}\,\Omega_{nm}(Z_0),$$
$$\mathrm{Im}\,\Omega_{nm}=\frac{(2f)^{(2n+1)/3}}{2^{2n}}\left(-\frac{F^2}{\omega^4}\right)^n\sum_{s=0}^{n}\frac{(-1)^s}{s!(2n+1-s)!}D_{nm}{}^s(Z_0),$$
$$D_{nm}^{(s)}(Z_0)=V^{(s+1)}V^{(2n+1-s)}-V^{(s)}V^{(2n+2-s)},\tag{35}$$
$$V^{(k)}=\frac{d^k}{dZ_0{}^k}V(Z_0).$$

When $n=1$, the expression given by (34) is equivalent to (14). The functions $\Omega_{nm}(Z_0)$ are nonzero for all values of the argument, i.e., in the presence of an electric field, the dissociating system can not only absorb but also emit an arbitrary number of photons ($n<m$). The field dependence of the threshold multiphoton EA probability follows directly from (35):

$$\Gamma_n(\omega=\alpha_F^2/2n,\ f)\sim f^{(2n+1)/3}.\tag{36}$$

According to (35), the dependence of n_f on the external electric field is determined by the three dimensionless parameters: F^2/ω^3, $f^{2/3}/\omega$, and Z_0.

Analysis of (35) shows that, as f increases in a weak electric field ($f^2/\omega^3<1$, $F^2/\omega^3<1$), the quantity n_f decreases, reaching a minimum for $f\sim\omega^{3/2}$ and begins to increase thereafter. This is illustrated in Fig. 3,

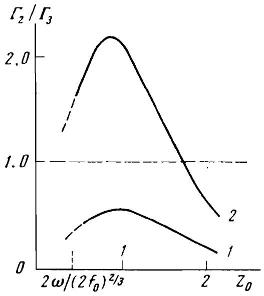

FIG. 3. Ratio of the probability of two-photon to three-photon ionization for the process (37) as a function of the parameter Z_0, calculated from (35). Curve 1—$F=10^7$ V/cm, curve 2—$F=5\times10^6$ V/cm.

which shows Γ_2/Γ_3 as a function of $Z_0=2\omega/(2f)^{2/3}$ for the process

$$\mathrm{Na}^- + n\omega \rightarrow \mathrm{Na} + e^- \qquad (37)$$

($|E_i|=3\omega$, $\omega=947$ cm^{-1}, which corresponds to the CO_2 laser radiation) for different wave fields. When $F\sim10^6$ V/cm, the increase in n_f occurs in an electric field f still weak enough in comparison with the characteristic atomic field $f_0=|E_i|^{3/2}$. The effect becomes more appreciable as the ratio $|E_i|/\omega$ increases. The nonmonotonic dependence of n_f on f will probably be found for neutral atoms as well.

We are indebted to V. S. Vinogradov for discussions of impurity EA in solids.

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1) We note that Prodan and Rozneritsa[15] considered two-photon EA, but their results are subject to computational errors.

2) It is well known that this quantity determines the van der Waals constants in systems containing the ion. When $f=0$, the formula given by (33) becomes identical with the expression for the polarizability of a negative ion given in[17].

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Optical orientation in ferromagnetic europium sulfide

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Results are presented of a study of optical spin orientation in a ferromagnetic semiconductor. The experimental results are interpreted in terms of a unified model which takes into account the effect of illumination on both the temperature and the Curie point of the spin system. Experiment shows that the Curie temperature of the magnetic semiconductor is raised by illumination.

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1. Optical spin orientation has by now been observed in a large number of paramagnetic semiconductors.[1] The effect of unpolarized light on a ferromagnetic medium has been investigated by several workers (see, for example,[2]). In this case, illumination gives rise to a change in the magnetic permeability of the medium. In contrast to previous experiments,[2] illumination by circularly polarized light produces optical magnetization of a demagnetized ferromagnet.[3,4] In this paper, we present the results of an investigation into the optical spin orientation in europium sulfide.

Europium sulfide is a magnetic semiconductor. Studies of the optical spin orientation in such compounds are of particular interest. From the standpoint of optical spin pumping, a ferromagnetic is the very opposite of a