Two-Raman-photon emission in an intense electromagnetic field

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Two-Raman-photon emission in hydrogen atoms in the presence of an intense electromagnetic field has been calculated utilizing the method of Dalgarno and Lewis. We give a general formulation for $(n-2)$-photon absorption and two-photon emission and also give numerical results for two-photon emission in the $1s$-$2p$ transition in hydrogen atoms. It is found that the emission of two spontaneous Raman photons could become important when the energy of the intense photon approaches the ionization energy. Finally, in the spectrum of the emitted Raman photon, faint lines are expected corresponding to the resonant peaks in the cross section.

I. INTRODUCTION

With the advent of the laser, very intense beams of photons have been developed so that processes involving emission or absorption of several photons can now be observed in the laboratory.[1-5] The interaction of such intense beams with atoms can lead to the emission of the electron from the atom, thus leading to the ionization of the gas and the excitation of the atom to some higher bound level.[6-18] It is also possible to study Raman-like processes in which a number of photons are absorbed and a single Raman photon is emitted. Theoretically, however, double-Raman-photon emission is also possible, though such an effect will be smaller than from a single-Raman-photon process. Simultaneous emission of two photons from the metastable $2s$ level of hydrogen was studied by Breit and Teller,[19] who also calculated the bounds for the lifetime of the $2s$ state in such a process. The two emitted photons were experimentally detected by Lipels *et al.*[20] in the decay of the metastable $^2S_{1/2}$ state of singly ionized helium, and the agreement with the theoretical result was good. In two-photon-emission processes the excitation energy is shared between the two photons, and therefore one gets a continuous band instead of a sharp line. The two-photon-continuum emission in deuterium-neon plasma has been detected by Elton *et al.*[21] By placing the atoms of the active medium in a suitable resonator, any portion of the above band can be amplified and a single sharp stimulated Raman line can be obtained.

In higher-order perturbation calculation, where many photon interactions are involved, there is the problem of summing over an infinite set of bound as well as continuum states of the unperturbed Hamiltonian. Some of the earlier calculations have either taken dominance of one term or utilized some sort of average to replace the infinite summation. A technique devised by Dalgarno and Lewis[22] and reformulated by Schwartz and Tieman[23] allows one to perform such a summation exactly. It was utilized by Mittleman and Wolf[24] to calculate the coherent scattering of photons by atomic hydrogen. Zernik[9] employed this technique for calculating two-photon ionization of the metastable level of hydrogen. The two-photon formulation of Zernik was generalized by Gontier and Trahin,[18] who presented numerical results for multiphoton ionization, bound-to-bound excitation, and single-Raman-photon emission.

In Sec. II we present the analytical formulation of our problem for two-Raman-photon emission and perform a separation between the angular variable and the radial matrix element, containing a sum over the infinite set of intermediate states.

In Sec. III we apply the technique of Dalgarno and Lewis,[22] as generalized by Gontier and Trahin[18] to sum over the intermediate states. The negative exponential present in the radial solution of the hydrogen atom is utilized to perform a Fourier transformation, leading to a first-order differential equation instead of a usual second-order one.

In Sec. IV we present the numerical results for the cross section. The transition matrix, as well as the total cross section, predicts resonant peaks whenever the energy of the emitted photon or that of intense photon corresponds to the energy difference of the bound levels. The total cross section increases rapidly as the energy of the intense photon approaches the ionization potential for the hydrogen atom, so that two-photon-emission process is likely to be important under such condition.

II. FORMULATION OF THE PROBLEM

The Hamiltonian of the hydrogen atom in the presence of a radiation field can be broken as usual into two parts—the unperturbed Hamiltonian H_0 and the interaction Hamiltonian H. The total Hamiltonian

$$H = H_0 + H_I ,$$

where

$$H_I = -(e/mc)\vec{A}\cdot\vec{p} + (e^2/2mc^2)A^2 ,$$

and e is the electronic charge, m the mass of the electron, p, its momentum, and c is the velocity of light. $\vec{A}$ is the vector potential of the external electromagnetic field. The term quadratic in $\vec{A}$ does not contribute to Raman-like processes, as has been shown by Cohen and Hameka,[25] and therefore in future consideration we shall always neglect it. The total cross section for the absorption of $(N-2)$ photons and emission of two Raman photons on the basis of Nth-order perturbation theory is given by

$$\sigma = 2r_0^2\alpha^3\left(\frac{I}{I_0}\right)^{N-3}\int_0^{E_m} E_\lambda{}^3 E_s E_s' \left|\frac{k^{(N)}}{E_\lambda{}^{N-1}}\right|^2_{\text{av}} dE_s , \quad (1)$$

where the average, av, is made over the direction of propagation and sum over polarization vectors of the two emitted photons, independently. In the right-hand side of the above expression, E_λ is the energy of the intense photon while E_s and E_s' are the energies of the two Raman photons. I stands for the flux of the incident radiation beam in W/cm^2 while $I_0 = 1.4038\times10^{17}$ W/cm^2; r_0 is the classical electron radius and α is the fine-structure constant. The energies of the photons and the matrix element K have been expressed in atomic units. As the conservation of energy holds, the energies of the emitted photons are related in the following way:

$$(N-2)E_\lambda - E_s - E_s' = E_f - E_g . \quad (2)$$

The maximum energy of one of the photons is

$$E_m = (N-2)E_\lambda - (E_f - E_g) , \quad (3)$$

E_f and E_g being the energies of the final and ground state, respectively. The Nth-order transition matrix is given by

$$k^{(N)} = \sum_{a_1,\ldots,a_{N-1}} \langle f|\vec{\epsilon}''\cdot\vec{p}|a_{N-2}\rangle \frac{\langle a_{N-1}|\vec{\epsilon}'\cdot\vec{p}|a_{N-2}\rangle}{E_{g,a_{N-1}}+(N-2)E_\lambda - E_s} \frac{\langle a_{N-2}|\vec{\epsilon}\cdot\vec{p}|a_{N-3}\rangle}{E_{g,a_{N-1}}+(N-2)E_\lambda}\cdots\frac{\langle a_1|\vec{\epsilon}\cdot\vec{p}|g\rangle}{E_{g,a_1}+E_\lambda} + (N^2-1)\text{ terms}, \quad (4)$$

containing all the permutations of $\vec{\epsilon}'$ and $\vec{\epsilon}''$. In the above equation $\vec{\epsilon}$ is the polarization vector of the incident electromagnetic wave; $\vec{\epsilon}'$ and $\vec{\epsilon}''$ are the polarization vectors of the spontaneously emitted Raman photons, and we have used the notation $E_{g,a_1} = E_g - E_{a_1}$. There are N^2 such terms, since $\vec{\epsilon}\cdot\vec{p}$ and $\vec{\epsilon}''\cdot\vec{p}$ can occur anywhere in the above expression. To simplify the notations of the expressions in the matrix element, we put

$$d_{k;\mu,\nu} = (\vec{\epsilon}\cdot\vec{p}) + \delta_{k,\mu}(\vec{\epsilon}'\cdot\vec{p} - \vec{\epsilon}\cdot\vec{p}) + \delta_{k,\nu}(\vec{\epsilon}''\cdot\vec{p} - \vec{\epsilon}\cdot\vec{p}) , \quad (5a)$$

$$\Omega_{k;\mu,\nu} = \Omega_{k-1;\mu,\nu} - \delta_{k,\mu}(E_s + E_\lambda) - \delta_{k,\nu}(E_s' + E_\lambda) + E_\lambda , \quad (5b)$$

with

$$\Omega_{0;\mu,\nu} = 0 . \quad (5c)$$

The first index denotes the position of the elements in the expression for the matrix element and the latter two indices give the positions of $\vec{\epsilon}'\cdot\vec{p}$ and $\vec{\epsilon}''\cdot\vec{p}$. With these notations the matrix element is expressed as a sum of components $M_{\mu,\nu}$ such that

$$k^{(N)} = \sum_{\mu,\nu} M_{\mu,\nu}^{(N)}$$

and

$$M_{\mu,\nu}^{(N)} = \sum_{a_1,\ldots,a_{N-1}} |\langle f|d_{N;\mu,\nu}|a_{N-1}\rangle \times \frac{\langle a_{N-1}|d_{N-1;\mu,\nu}|a_{N-2}\rangle}{E_{g,a_{N-1}}+\Omega_{N-1;\mu,\nu}}\cdots\frac{\langle a_1|d|g\rangle}{E_{g\mu_1}+\Omega_{1;\mu\nu}} . \quad (6)$$

To simplify $M_{\mu,\nu}$ further, we take the direction of the incident electromagnetic polarization vector to be along the z direction so that $\vec{\epsilon}\cdot\vec{p} = p_z$. The polarization vector of the emitted photons can be taken outside and we get

$$M_{\mu,\nu} = \left(\frac{\vec{\epsilon}\cdot\vec{\epsilon}'}{3}\right)\left(\frac{\vec{\epsilon}\cdot\vec{\epsilon}''}{3}\right)\sum_{a_1,\ldots,a_{N-1}} \langle f|p_z|a_{N-1}\rangle \frac{\langle a_{N-1}|p_z|a_{N-2}\rangle}{E_{g,a_{N-1}}+\Omega_{N-1;\mu,\nu}} \frac{\langle a_\mu|\vec{p}|a_{\mu-1}\rangle}{E_{g,a_\mu}+\Omega_{\mu;\mu,\nu}}\cdot\frac{\langle a_{\mu-1}|\vec{p}|a_{\mu-2}\rangle}{E_{g,a_{\mu-1}}+\Omega_{\mu-1;\mu,\nu}}\cdots$$

$$\times\frac{\langle a_\nu|\vec{p}|a_{\nu-1}\rangle}{E_{g,a_\nu}+\Omega_{\nu;\mu,\nu}}\cdot\frac{\langle a_{\nu-1}|\vec{p}|a_{\nu-2}\rangle}{E_{g,a_{\nu-1}}+\Omega_{\nu-1;\mu,\nu}}\cdots\frac{\langle a_1|p_z|g\rangle}{E_{g,a_1}+\Omega_{1;\mu,\nu}} . \quad (7)$$

The summation over complete sets of states is made of angular as well as radial parts. The summation over the angular variables can be performed if we take help of the following relations:

$$\nabla_z[R_{n,l}(r)Y_l^m(\theta,\phi)]=[A_{l,m}Y_{l+1}^m(\theta,\phi)q^+ + B_{l,m}Y_{l+1}^m(\theta,\phi)q^-]R_{n,l}(r), \tag{8a}$$

$$(\nabla_\kappa + i\nabla_y)[R_{n,l}(r)Y_l^m(\theta,\phi)]=[C_{l,m}Y_{l+1}^{m+1}(\theta,\phi)q^+ + D_{l,m}Y_{l-1}^{m+1}(\theta,\phi)q^-]R_{n,l}(r), \tag{8b}$$

$$(\nabla_\kappa - i\nabla_y)[R_{n,l}(r)Y_l^m(\theta,\phi)]=[E_{l,m}Y_{l+1}^{m-1}(\theta,\phi)q^+ + F_{l+1}^{m-1}(\theta,\phi)q^-]R_{n,l}(r). \tag{8c}$$

The constants $A_{l,m}$, $B_{l,m}$, $C_{l,m}$, $D_{l,m}$, $E_{l,m}$, $F_{l,m}$ are functions of the quantum number l and m and are tabulated in Ref. 26. The operators $q^\pm$ are given by

$$q^+(l,r)=\frac{d}{dr}-\frac{l}{r}, \tag{9}$$

$$q^-(l,r)=\frac{d}{dr}+\frac{l+1}{r}. \tag{10}$$

Since the angular momentum of the two states in the matrix element $\langle\psi_{j+1}|\nabla^{\pm,z}|\psi_j\rangle$ differ by ± 1 a convenient expression for it is

$$q(l_j,r)=\frac{l_{j+1}-l_j+1}{2}q^+(l_j,r) + \frac{l_j-l_{j+1}+1}{2}q^-(l_j,r). \tag{11}$$

If we denote the hydrogenic-atom wave functions by $\psi_{n_j,l_j,m_j}\equiv R_{l_j}Y_{l_j}^{m_j}$, where R_{l_j} is the radial part, we then get

$$\langle\psi_{l_{j+1}}|\nabla_z|\psi_{l_j}\rangle=Q^0(l_{j+1};l_j)\times\langle R_{l_{j+1}}|q|R_{l_j}\rangle, \tag{12a}$$

$$\langle\psi_{l_{j+1}}|\nabla_\kappa+i\nabla_y|\psi_{l_j}\rangle=Q^+(l_j+1,l_j)\times\langle R_{l_{j+1}}|q|R_{l_j}\rangle, \tag{12b}$$

$$\langle\psi_{l_{j+1}}|\nabla_\kappa-i\nabla_y|\psi_{l_j}\rangle=Q^-(l_j+1,l_j)\times\langle R_{l_{j+1}}|q|R_{l_j}\rangle, \tag{12c}$$

where we have put

$$Q^0(l_j+1,l_j)=A_{l_j,m_j}\delta(l_{j+1},l_j+1)+B_{l_j,m_j}\delta(l_{j+1},l_j-1), \tag{13a}$$

$$Q^+(l_{j+1},l_j)=C_{l_j,m_j}\delta(l_{j+1}l_j+1)+D_{l_j,m_j}\delta(l_{j+1},l_j-1), \tag{13b}$$

$$Q^-(l_{j+1},l_j)=E_{l_j,m_j}\delta(l_{j+1},l_j+1)+F_{l_j,m_j}\delta(l_{j+1},l_j-1). \tag{13c}$$

Using Eqs. (12) and (13) we can write $M_{\mu,\nu}$ as

$$M_{\mu,\nu}=G_{\mu,\nu}T. \tag{14}$$

Here $G_{\mu,\nu}$ contains all the angular contributions and T contains the radial part and is given by (the subscripts μ, ν on T is dropped)

$$T=\sum_{a_1,\ldots,a_{N-1}}\langle f|q|a_{N-1}\rangle\times\frac{\langle a_{N-1}|q|a_{N-2}\rangle}{E_{g,a_{N-1}}+\Omega_{N-1;\mu,\nu}}\cdots\frac{\langle a_1|q|g\rangle}{E_{g,a_1}+\Omega_{1;\mu,\nu}}. \tag{15}$$

The angular part is given by

$$\begin{aligned}G_{\mu,\nu}=&\left(\frac{\vec\epsilon\cdot\vec\epsilon\,'}{3}\right)\left(\frac{\vec\epsilon\cdot\vec\epsilon\,''}{3}\right)Q^0(l_N,l_{N-1})Q^0(l_{N-1},l_{N-2})\cdots\\&\times\{Q^0(l_\mu,l_{\mu-1})Q^0(l_{\mu-1},l_{\mu-2})+\tfrac{1}{2}[Q^+(l_\mu,l_{\mu-1})Q^-(l_{\mu-1},l_{\mu-2})+Q^-(l_\mu,l_{\mu-1})Q^+(l_{\mu-1},l_{\mu-2})]\}\\&\times Q^0(l_{\mu-2},l_{\mu-3})\cdots\{Q^0(l_\nu,l_{\nu-1})Q^0(l_{\nu-1},l_{\nu-2})+\tfrac{1}{2}[Q^+(l_\nu,l_{\nu-1})Q^-(l_{\nu-1},l_{\nu-2})+Q^-(l_\nu,l_{\nu-1})Q^+(l_{\nu-1},l_{\nu-2})]\}\\&\times Q^0(l_{\nu-2},l_{\nu-3})\cdots Q_0(l_1,l_g).\end{aligned} \tag{16}$$

Further

$$G_{\mu,\nu}=\left(\frac{\vec\epsilon\cdot\vec\epsilon\,'}{3}\right)\left(\frac{\vec\epsilon\cdot\vec\epsilon\,''}{3}\right)\bar G_{\mu,\nu}.$$

Summing over the polarizations and averaging over directions of the emitted photons, we have

$$|K^{(N)}|^2_{\rm av}=\left(\frac{2}{27}\right)^2\left|\sum_{\mu,\nu}G_{\mu,\nu}(l_1,m_1;l_2,m_2;\ldots;L,M)\,T(l_1,l_2,\ldots,L|E_\lambda,E_s,E_s')\right|^2. \tag{17}$$

III. SUMMATION TECHNIQUE

We now discuss how to perform the intermediate-states summation occurring in Eq. (15). To perform the summation over the complete set of radial states, we define function V, which is closely related to the

T matrix. For example, we define

$$T(l, l_1, \ldots, L \mid E_\lambda, E_s, E_s') = \int_0^\infty dr\, r^2 R_{n,l}(r) q(l,r)\, V(l_1, l_2, \ldots, L \mid r, E_\lambda, E_s, E_s') . \tag{18}$$

The $R_{n,l}(r)$ are the radial wave functions of the hydrogen ground state, and V is given by

$$V(l_1, l_2, \ldots, L \mid \gamma, E_\lambda, E_s, E_s') = \sum_{n_1, \ldots, n_{N-1}} R_{n_1 L_1}(r) \frac{\langle R_{n_1} | q | R_2 \rangle}{E_{g,n_1} + \Omega_{n_1;\mu,\nu}} \cdots \frac{\langle R_{N-1} | q | R_f \rangle}{E_{g,N-1} + \Omega_{N-1;\mu,\nu}} . \tag{19}$$

The sequence of the V's of different order satisfies

$$V(l_j, l_{j+1}, \ldots, L \mid \gamma, E_\lambda, E_s, E_s') = \sum_{nj} R_{n_j, l_j}(r_j) \int_0^\infty dr_j r_j{}^2 R_{n_j, l_j}(r_j) \frac{1}{E_{g2_j} + \Omega_{j;\mu,\nu}}$$
$$\times q(l_j + 1, r_j)\, V(l_{j+1}, l_{j+2}, \ldots, L \mid \gamma, E_\lambda, E_s, E_s')\quad V(L \mid \gamma, E_\lambda) = R_{n_f, l_f}(r) . \tag{20}$$

Since $R_{n_j, l_j}(r)$ is a solution of the radial equation for the hydrogen atom, it must satisfy

$$D_{l_j}(r) R_{n_j, l_j}(v) \equiv \left(\frac{1}{2} \frac{d^2}{dr^2} + \frac{1}{r} \frac{d}{dv} + \frac{1}{r} - \frac{l_j(l_j+1)}{2r^2} \right) R_{n_j, l_j}(r)$$
$$= -E_{n_j} R_{n_j, l_j}(r) . \tag{21}$$

Equation (21) can be used further to simplify Eq. (20), and we get

$$(E_g + \Omega_{j;\mu,\nu} + D_{l_j})\, V(l_j, l_{j+1}, \ldots, L \mid r, E_\lambda, E_s, E_s')$$
$$= \int_0^\infty dr_j (r_j / r) \sum_{n_j, l_j} r R_{n_j, l_j}(r_j)$$
$$\times q(l_j + 1, \gamma) V(l_{j+1}, l_{j+2}, \ldots, L \mid r, E_\lambda, E_s, E_s') . \tag{22}$$

The sequence of the V's are therefore related by the following second-order differential equation:

$$(E_g + \Omega_{j;\mu,\nu} + D_{l_j})\, V(l_j, \ldots, L \mid \gamma, E_\lambda, E_s, E_s')$$
$$= q(l_j + 1, \gamma) V(l_{j+1}, \ldots, L \mid r, E_\lambda, E_s, E_s') . \tag{23}$$

Let us now define a function $y(l_1, \ldots, L \mid p)$ by the following:

$$y(l_j, l_{j+1}, \ldots, L \mid p) = \left(\frac{d}{dp}\right)^{l_{j+1}} \int_0^\infty dr\, e^{-pr}$$
$$\times V(l_j, l_{j+1}, \ldots, L/\gamma, E_\lambda, E_s E_s')$$
$$= (-1)^{l_j+1} \int_0^\infty dr\, e^{-pr} r^{l_j+1}$$
$$\times V(l_j, l_{j+1}, \ldots, L \mid r, E_\lambda, E_s, E_s') . \tag{24}$$

By using Eq. (24), Eq. (23) reduces to the following first-order differential equation:

$$\left((p^2 - \alpha_j{}^2) \frac{d}{dp} + 2[(l_{j+1})p - 1] \right) y(l_j, l_{j+1}, \ldots, L \mid p)$$
$$= \left[\delta(l_{j+1}, l_j - 1) \left(2p \frac{d^2}{dp^2} + (4 l_j + 2) \frac{d}{dp} \right) + \delta(l_{j+1}, l_j + 1) 2p \right] y(l_{j+1}, \ldots, L \mid p), \tag{25}$$

where

$$\alpha_j^2 = -2(E_g + \Omega_{j;\mu,\nu}) .$$

Equation (25) can be used successively to obtain all the functions y. For example, from the initial given function $y(L|p)$ we get for $y(l_{N-1}, L|p)$ the following relations:

$$\left((p^2 - \alpha_{N-1;\mu,\nu}^2) \frac{d}{dp} + 2[(l_{N-1} + 1)p - 1] \right) y(l_{N-1}, L \mid p)$$
$$= \left[\delta(L, l_{N-1} - 1) \left(2p \frac{d^2}{dp^2} + (4 l_{N-1} + 2) \frac{d}{dp} \right) + \delta(L, l_{N-1} + 1) 2p \right] y(L \mid p) \tag{26}$$

and

$$y(L \mid p) = \frac{d^{L+1}}{dp^{L+1}} \int_0^\infty dr\, R_{n,L}(r)\, e^{-pr} .$$

Thus, in principle, one can obtain the cross section if one is able to solve successively all the first-order differential equations given by Eq. (25). We have solved the above equations by a Taylor-series expansion method, which has been discussed thoroughly by Zernik and Klopfenstein in Ref. 9. For the transition from the ground state the transition matrix $T(l, l_1, \ldots, L \mid \gamma, E_\lambda, E_s, E_s')$

is related to $y(l_1, l_2, \ldots, L|p)$ in the following way:

$$T(l, l_1, \ldots, L|E_\lambda, E_s, E_s') = 2\left[\delta(l, l_1+1)\left(p\frac{d^2}{dp^2}+\frac{d}{dp}\right)+\delta(l, l_1-1)p\right]y(l_1, \ldots, L|p))|_{p=1/n}\,. \tag{27}$$

Equations (1) and (17) can now be used in conjunction with Eq. (27) to obtain two-photon-emission cross section.

Using the above result, we have calculated the two-photon-emission cross section in the transition $1s$-$2p$ by a numerical procedure. In the lowest order, the electron absorbs one intense photon in the ground state and is thereby excited. It then goes to the $2p$ state with the simultaneous emission of two Raman photons. In our case of one-intense-photon absorption and two-Raman-photon emission, we have to solve two sets of coupled first-order differential equations. Our numerical results are present in Figs. 1 and 2. Recently a number of workers[27] have found the amplification by the stimulated emission of radiation at photon energies above 10 eV, so that the experiment in the ground state of hydrogen could be performed in the near future.

IV. DISCUSSION

The matrix element $K^{(N)}$ as a function of the energy of one of the emitted photons shows resonant peaks whenever its energy becomes equal to the difference between the two bound levels (Fig. 1). These resonant peaks could manifest themselves as faint lines in the emission spectra. Another interesting property is that (if we average-out the resonant peaks) the matrix element decreases both for high and low values of the emitted photon energy, i.e., the two photons tend to have nearly equal energies. A similar result has been derived, by Elton *et al.*,[21] for two-photon emission from the excited state of helium and has been experimentally verified by Spitzer and Greenstein[28] in helium-neon. The numerical value for the cross section has been plotted against the energy of the intense photon in Fig. 2. The cross section exhibits the usual resonant character whenever the energy of the intense photon corresponds to the energy difference of the bound levels of hydrogen. An important feature of the curve is the increase in the cross section by several orders as the intense photon energy increases, making it a dominant process in the vicinity of ionization. The result is similar to one found by Mohan and Thareja[17] that two-photon-induced emission is enhanced over one-photon emission. In fact, for the values of the intensity of laser beam of the order of 10^6 W/cm^2, two-photon emission could exceed ionization. This is not surprising, as in similar conditions the cross section for a bound-to-bound transition with single-Raman-photon emission also exceeds the ionization cross section (Gontier and Trahin,[18] 1971). Therefore, in conclusion, we can say that the emission of two spontaneous Raman photons is likely to become important for the energy of the intense photon approach-

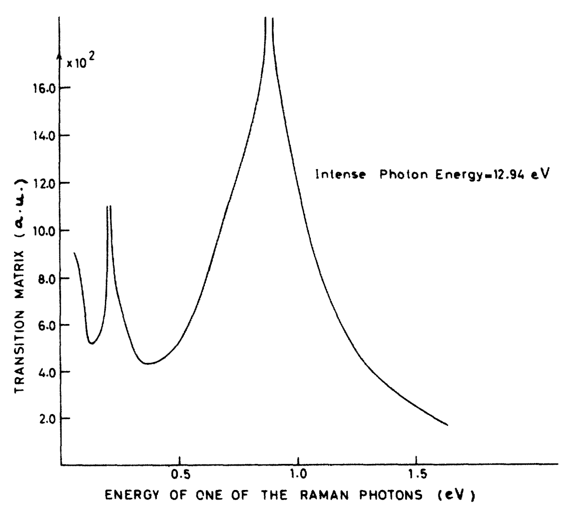

FIG. 1. Transition matrix as a function of the energy of one of the emitted Raman photons.

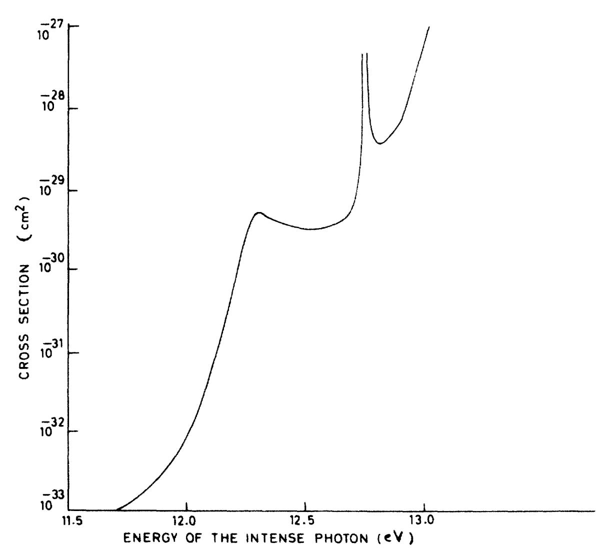

FIG. 2. Cross section as a function of the energy of the intense photon.

ing the ionization energy. Also, in the spectrum of the emitted Raman photon we may expect faint lines corresponding to the resonant peaks in the cross section and transition matrix.

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