to the field, and ρ the magnitude of the orthogonal coordinate. We take for β the fixed state const.$\times B^{1/2} \times e^{-B\rho^2/4}$ and vary α. One then obtains the variational problem for a one-dimensional potential $W(z)$ const. $\times B \int V(\rho, \varphi, z) e^{-B\rho^2/2} \rho d\rho d\varphi$ so that small B corresponds to weak W where the binding energy [see B. Simon, Ann. Phys. (N.Y.) 97, 279 (1976)] is $[\frac{1}{2}\int W(z)\, dz]^2$.

[13] J. J. Hopfield, P. J. Dean, and D. G. Thomas, Phys. Rev. 158, 748–755 (1967).

[14] The inequality (2) comes from the following: The lowest state for which the electron can become unbound has an energy $B/2m$ due to the zero-point energy of the Landau oscillator. On the other hand, the energy of the ground state must go up in a magnetic field [see B. Simon, Phys. Rev. Lett. 36, 1083–1084 (1976)] and we are supposing it to be zero if $B=0$.

[15] If $V(r) = -\alpha/(\beta + r^2)$, with $0 \leqslant 2m\alpha < 1/4$ (which implies no bound state if $B=0$), one finds by an explicit variational computation that

$$\lim_{B \downarrow 0} E(B)/B \geqslant \pi^3 \alpha^2 m$$

which can be made arbitrarily close to $(\pi^3/32)(\frac{1}{2}m) = (0.969)(\frac{1}{2}m)$. This comes close to saturating the bound (2).

Choice of Gauge in Two-Photon Transitions: 1*s*-2*s* Transition in Atomic Hydrogen

F. Bassani
Istituto di Fisica, Università di Roma, Roma, Italy

and

J. J. Forney and A. Quattropani
Laboratoire de Physique Théorique, Ecole Polytechnique Fédérale, Lausanne, Switzerland

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The two-photon transition rate for hydrogen is evaluated with the $\vec{E}\cdot\vec{x}$ and the $\vec{A}\cdot\vec{p}$ interactions including the continuum. We find that the $\vec{E}\cdot\vec{x}$ interaction approaches the final result with a small number of intermediate states and consequently is the one to be used in any approximate calculation. Numerical values of the two-photon absorption coefficient as a function of frequency are given and the resonance enhancement is exhibited.

The problem of the choice of the gauge in multiphoton transitions has been discussed by a number of authors[1,2] in connection with the proper choice of the unperturbed wave functions required to insure gauge invariance. We have shown[2] that, at least, in the dipole approximation the two expressions $\vec{E}\cdot\vec{x}$ and $\vec{A}\cdot\vec{p}$ for the photon-electron interaction give the same transition probability to all orders with the same unperturbed wave functions, provided that the complete set of intermediate states is used. In an exact calculation it is thus irrelevant to use one or the other form of the interaction. However, in general only a limited number of intermediate states can be taken into account and the problem of which interaction gives more reliable results in this approximation is essential.

In order to answer this question we have considered a case which can be solved exactly, the two-photon 1*s*-2*s* transition in hydrogen. We present the exact results for the transition rate and absorption coefficient in the two gauges and we compare the convergence over all intermediate states including the continuum. The result of our analysis indicates that the $\vec{E}\cdot\vec{x}$ interaction is the one to be used in any approximate calculation.

The interaction of radiation with the electron in the two gauges, denoted by J_0 and J, takes the following forms in mks units:

$$V^{J_0} = -eE_1\hat{e}_1\cdot\vec{r}\cos\omega_1 t - eE_2\hat{e}_2\cdot\vec{r}\cos\omega_2 t, \tag{1a}$$

$$V^{J} = eE_1\frac{\hat{e}_1\cdot\vec{p}}{m\omega_1}\sin\omega_1 t + eE_2\frac{\hat{e}_2\cdot\vec{p}}{m\omega_2}\sin\omega_2 t, \tag{1b}$$

where E_1 and E_2 are the electric fields of the incident electromagnetic waves traveling in the y direction with polarization vectors $\hat{e}_1$ and $\hat{e}_2$ and frequencies ω_1 and ω_2.

The two-photon transition probability per unit time and per atom between the 1*s* and 2*s* states reads

$$W(1s\text{-}2s) = \frac{|E_1E_2|^2 e^4 a_0^4}{4(2\pi\hbar^2)^2 \mathfrak{R}^3}\,\tfrac{1}{9}\,|D[J_0]|^2\delta(\Delta\nu), \tag{2a}$$

$$W(1s\text{-}2s) = \frac{|E_1E_2|^2 e^4 a_0^4}{4(2\pi\hbar^2)^2 \mathfrak{R}^3}\,\tfrac{1}{9}\,|D[J]|^2\delta(\Delta\nu), \tag{2b}$$

in the gauges J_0 and J, respectively. In (2), a_0 denotes the Bohr radius; $\mathfrak{R} = 3.29\times 10^{15}$ sec^{-1}, the Rydberg frequency; and

$$D[J_0] = \frac{3}{2}\sum (1+P_{12}) \frac{\hat{e}_1 \cdot \langle 2s|\vec{\mathrm{r}}/a_0|n\rangle\langle n|\vec{\mathrm{r}}/a_0|1s\rangle \cdot \hat{e}_2}{\nu(n) - \nu(1s) - \nu_2}, \tag{3a}$$

$$D[J] = \frac{3}{2}\sum (1+P_{12}) \frac{\hat{e}_1 \cdot \langle 2s|\vec{\mathrm{p}}/m\omega_1 a_0|n\rangle\langle n|\vec{\mathrm{p}}/m\omega_2 a_0|1s\rangle \cdot \hat{e}_2}{\nu(n) - \nu(1s) - \nu_2}, \tag{3b}$$

where P_{12} interchanges the frequencies and polarization of the two photons. The sums in (3) extend over the discrete and continuum spectrum; ν denotes dimensionless frequencies defined as

$$\omega_i = 2\pi\mathcal{R}\nu_i, \quad \nu(1s) = E_{1s}/2\pi\hbar\mathcal{R} = -1, \quad \nu(2s) = E_{2s}/2\pi\hbar\mathcal{R} = -\tfrac{1}{4};$$

and $\nu(n)$ are the frequencies of the intermediate states $|n\rangle$. The argument of the energy-conserving δ function reads $\Delta\nu = \nu(2s) - \nu(1s) - \nu_1 - \nu_2$.

Using the well-known relation $\vec{\mathrm{p}} = -(im/\hbar)[\vec{\mathrm{r}}, H_0]$, we transform the matrix elements of $\vec{\mathrm{p}}$ into matrix elements of $\vec{\mathrm{r}}$: $\langle n'|\vec{\mathrm{p}}|n\rangle = \langle n'|\vec{\mathrm{r}}|n\rangle(E_{n'} - E_n)im/\hbar$. This relation is also valid for a many-electron system if one uses exact eigenstates, but has to be corrected as indicated by Starace[3] if one uses exact eigenstates of an approximate Hamiltonian which contains nonlocal potentials as, for instance, Hartree-Fock eigenstates. The explicit expression for D in both gauges is given for the discrete spectrum by

$$D^d[J_0] = \lim_{N\to\infty} \frac{1}{2}\sum_{n=2}^{N} \left\{ \left(1 - \frac{1}{n^2} - \nu_1\right)^{-1} + \left(\frac{1}{4} - \frac{1}{n^2} + \nu_1\right)^{-1} \right\} R_{10}{}^{n1} R_{20}{}^{n1}, \tag{4a}$$

$$D^d[J] = \lim_{N\to\infty} -\frac{1}{2}\sum_{n=2}^{N} \left\{ \left(1 - \frac{1}{n^2} - \nu_1\right)^{-1} + \left(\frac{1}{4} - \frac{1}{n^2} + \nu_1\right)^{-1} \right\} R_{10}{}^{n1} R_{20}{}^{n1} \left(\frac{1}{4} - \frac{1}{n^2}\right)\left(1 - \frac{1}{n^2}\right)\left[\nu_1\left(\frac{1}{4} - \nu_1\right)\right]^{-1}, \tag{4b}$$

TABLE I. Dimensionless two-photon transition probability amplitude $D[J_0]$ for $1s$-$2s$ transitions in hydrogen as a function of the incident photon frequency ν_1 in the gauge $\vec{\mathrm{E}}\cdot\vec{\mathrm{r}}$. The argument N denotes the highest intermediate state included in (4a). $D^d[J_0N]$ denotes the contribution from the discrete spectrum up to the Nth intermediate state. $D^c[J_0]$ denotes the contribution from the continuum.

ν_1 $D^d[J_0N]$	0.3750	0.5250	0.6750	0.6875	0.7000	0.7125	0.7250	0.7375	0.7475
N= 2	-17.8785	-21.2839	-49.6624	-58.5113	-71.8331	-94.0971	-138.712	-272.722	-1345.37
3	-14.7969	-17.9154	-44.9877	-53.6216	-66.6974	-88.6783	-132.964	-266.589	-1338.87
4	-14.1026	-17.1679	-44.0174	-52.6173	-65.6552	-87.5934	-131.831	-265.401	-1337.64
5	-13.8265	-16.8723	-43.6427	-52.2309	-65.2558	-87.1795	-131.401	-264.953	-1337.17
6	-13.6861	-16.7225	-43.4550	-52.0376	-65.0564	-86.9733	-131.187	-264.731	-1336.94
7	-13.6042	-16.6352	-43.3463	-51.9258	-64.9411	-86.8543	-131.064	-264.603	-1336.81
8	-13.5519	-16.5795	-43.2773	-51.8548	-64.8680	-86.7788	-130.986	-264.522	-1336.73
9	-13.5163	-16.5417	-43.2305	-51.8068	-64.8186	-86.7278	-130.933	-264.467	-1336.67
10	-13.4910	-16.5148	-43.1973	-51.7727	-64.7834	-86.6916	-130.896	-264.428	-1336.63
11	-13.4723	-16.4949	-43.1728	-51.7475	-64.7575	-86.6649	-130.868	-264.400	-1336.60
12	-13.4581	-16.4798	-43.1542	-51.7284	-64.7379	-86.6446	-130.847	-264.378	-1336.58
13	-13.4470	-16.4680	-43.1397	-51.7135	-64.7226	-86.6288	-130.831	-264.361	-1336.56
∞	-13.3824	-16.3994	-43.0555	-51.6271	-64.6337	-86.5373	-130.737	-264.263	-1336.46
$D^c[J_0]$	1.6019	1.6675	1.9071	1.9393	1.9742	2.0121	2.053	2.098	2.14
$D[J_0]$	-11.7805	-14.7319	-41.1484	-49.6878	-62.6595	-84.5252	-128.683	-262.165	-1334.33

TABLE II. Dimensionless two-photon transition probability amplitude $D[J]$ for $1s$-$2s$ transitions in hydrogen as a function of the incident photon frequency ν_1 in the gauge $\vec{A}\cdot\vec{p}$. The argument N denotes the highest intermediate state included in (4b). $D^d[JN]$ denotes the contribution from the discrete spectrum up to the Nth intermediate state. $D^c[J]$ denotes the contribution from the continuum.

$D^d[JN]$ \ ν_1	0.3750	0.5250	0.6750	0.6875	0.7000	0.7125	0.7250	0.7375	0.7475
N= 2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	-2.7053	-3.5206	-11.3999	-14.0490	-18.1150	-25.0382	-39.1501	- 82.1401	-428.970
4	-3.5732	-4.6329	-14.7691	-18.1575	-23.3496	-32.1756	-50.1379	-104.783	-545.294
5	-3.9691	-5.1373	-16.2610	-19.9702	-25.6500	-35.2983	-54.9213	-114.585	-595.394
6	-4.1847	-5.4113	-17.0622	-20.9421	-26.8811	-36.9659	-57.4697	-119.794	-621.955
7	-4.3158	-5.5775	-17.5451	-21.5273	-27.6216	-37.9679	-58.9990	-122.915	-637.851
8	-4.4016	-5.6863	-17.8597	-21.9084	-28.1034	-38.6193	-59.9925	-124.941	-648.161
9	-4.4609	-5.7614	-18.0766	-22.1709	-28.4352	-39.0677	-60.6760	-126.334	-655.245
10	-4.5037	-5.8156	-18.2325	-22.3597	-28.6737	-39.3899	-61.1670	-127.334	-660.330
11	-4.5356	-5.8559	-18.3485	-22.5000	-28.8511	-39.6295	-61.5319	-128.077	-664.107
12	-4.5600	-5.8868	-18.4372	-22.6074	-28.9866	-39.8125	-61.8106	-128.644	-666.992
13	-4.5791	-5.9110	-18.5066	-22.6913	-29.0926	-39.9556	-62.0285	-129.088	-669.245
∞	-4.6924	-6.0542	-18.9167	-23.1872	-29.7187	-40.8006	-63.3146	-131.704	-682.536
$D^c[J]$	-7.0881	-8.6777	-22.2317	-26.5005	-32.9408	-43.7245	-65.3689	-130.461	-651.790
$D[J]$	-11.7805	-14.7319	-41.1484	-49.6877	-62.6595	-84.5252	-128.683	-262.165	-1334.33

and for the continuum by

$$D^c[J_0]=\tfrac{3}{2}\int_0^\infty dw\,\{(2w+1-\nu_1)^{-1}+(2w+\tfrac{1}{4}+\nu_1)^{-1}\}X_{w1}X_{w2}, \tag{5a}$$

$$D^c[J]=-\tfrac{3}{2}\int_0^\infty dw\,\{(2w+1-\nu_1)^{-1}+(2w+\tfrac{1}{4}+\nu_1)^{-1}\}X_{w1}X_{w2}(\tfrac{1}{4}+2w)(2w+1)[\nu_1(\tfrac{3}{4}-\nu)]^{-1}. \tag{5b}$$

The matrix elements of $|r|$ between the s states and the p_z states of the discrete and continuum spectra are taken from Bethe and Salpeter,[4] Eqs. (63.4), (71.4), and (71.14). We notice that all matrix elements are positive with the exception of $R_{20}{}^{21}=-\sqrt{27}$. The above expressions (4) and (5) refer to photons polarized in the same direction; for polarization angle θ between the two photons, expressions (4) and (5) have to be multiplied by $\cos\theta$.

The evaluation of (4) and (5) is performed numerically and the results are given in Tables I and II for the gauges J_0 and J, respectively. We notice that both gauges give exactly the same final result, but the respective contributions from intermediate states are dramatically different. In particular the $2p$ state (degenerate with the final state $2s$) gives the dominant contribution in the $\vec{E}\cdot\vec{r}$ gauge, while it gives contribution zero in the $\vec{A}\cdot\vec{p}$ gauge. It can also be observed that the continuum spectrum gives a negligible contribution in the $\vec{E}\cdot\vec{r}$ gauge, while it gives more than half the total contribution in the $\vec{A}\cdot\vec{p}$ gauge.

From the above results one easily obtains the absorption coefficient for photons of frequency ω_1 in the presence of N_2 photons per unit volume of frequency ω_2. The absorption coefficient is given in terms of the transition rate by

$$\alpha(\omega_1)=\frac{W(1s-2s)\hbar\omega_1 N_0}{\frac{1}{2}\epsilon_1\epsilon_0|E_1|^2 c/\sqrt{\epsilon_1}}, \tag{6}$$

where N_0 is the density of atoms in the sample and ϵ_1 is the dielectric function of the medium for the radiation of frequency ω_1. The density of photons of frequency ω_2 is related to the energy density by

$$\tfrac{1}{2}\epsilon_0\epsilon_2|E_2|^2=\hbar\omega_2 N_2.$$

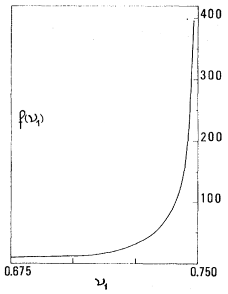

FIG. 1. Dimensionless absorption coefficient $f(\nu_1)$ as a function of the incident photon frequency ν_1. The variation of $f(\nu_1)$ for $0.3750 < \nu_1 < 0.6750$ is negligible.

Introducing (2) and (7) into (6) we obtain

$$\alpha(\omega_1) = \frac{e^4 a_0{}^4}{\epsilon_0{}^2 \hbar^2 \Re c} \frac{N_2 N_0}{\epsilon^2 \sqrt{\epsilon_1}} \nu_1 \nu_2 \frac{1}{g} |D|^2 \delta(\Delta\nu) \tag{7}$$

$$= \frac{e^4 a_0{}^4}{\epsilon_0{}^2 \hbar^2 \Re c} \frac{N_2 N_0}{\epsilon_2 \sqrt{\epsilon_1}} f(\nu_1) \tag{8}$$

where $f(\nu_1)$ is a dimensionless function. From (8) we can also obtain the emission coefficient and the natural lifetime of the metastable state $2s$, which has been previously computed.[5]

The function $f(\nu_1)$ is plotted in Fig. 1, which clearly shows the resonant enhancement as ν_1 approaches the $1s$-$2p$ separation. Such an effect has been observed for the $3s$-$4d$ transition of sodium vapor.[6] The $1s$-$2s$ transition in hydrogen has been observed, but the value of the absorption is not given.[7]

The results here presented allow us to draw some conclusions which go beyond the specific case of hydrogen. First of all, in order to obtain a gauge-invariant two-photon transition rate one has to include the complete set of intermediate states, which is in general not feasible. The convergence of the transition rate on the intermediate states depends dramatically on the choice of the gauge. Since a much better convergence is obtained in the preferential gauge $\vec{E}\cdot\vec{x}$ we conclude that this is the interaction to be adopted.

The reason for such results may be intuitively understood by considering that the connection between matrix elements of $\vec{p}$ and of $\vec{r}$ introduces in Eq. (3b) the factor $(E_{2s}-E_n)(E_n-E_{1s})/\hbar^2\omega_1\omega_2$ which becomes very large for E_n very far from initial or final states and consequently enhances the contributions of those intermediate states in the gauge $\vec{A}\cdot\vec{p}$. Similar considerations were brought forward by Dalgarno and Lewis[8] for one-photon transitions, when initial and final wave functions are expanded in a particular basis.

Our result is not confined to atoms but can be extended to electronic transitions in crystals, where in general only one intermediate state is considered. In particular, our result, showing that the intermediate state which lies nearest to the final state is the dominant one for the $\vec{E}\cdot\vec{x}$ interaction, justifies the validity of the Mahan model with respect to the Loudon model for two-photon production of excitons.[9]

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