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Effect of Intensity on Multiphoton Ionization

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For multiphoton processes, the higher-order terms of the perturbation series are treated by summing the class of irreducible diagrams. This allows the examination of the behavior of the cross section as a function of the incident photon flux at arbitrarily large intensities by means of analytic continuation to the nonperturbative region. The case of four-photon ionization of the ground state of hydrogen is discussed as an example.

In recent years, there has been a great deal of interest in multiphoton processes. While an N-photon process is basically well described by the Nth-order term of the perturbation series for the transition amplitude, there are two cases where the contributions of the higher-order terms need to be taken into account. Firstly, the intensity of the external field may be large enough to render significant contributions from the higher-order terms, and secondly, at frequencies close to resonances,[1,2] the contributions of the higher-order terms become very important.

In this Letter, we report progress in understanding the contributions of the higher-order terms of the perturbation series. Our main results are the following: (1) We have developed an algorithm to sum the entire perturbation series for an N-photon process as represented by the irreducible Feynman diagrams. (Irreducible diagrams are those which cannot be cut to produce an N-photon absorption graph.) (2) The initial and the final states must be renormalized and this takes into account all the reducible diagrams. Our result, therefore, contains all terms of the perturbation series arising from the electromagnetic field in the dipole approximation interacting with a one-electron atom. (3) The transition amplitude can be analytically continued to regions of very large external field where the perturbation series ceases to be meaningful.

We illustrate our method for the case of $N=4$ (four-photon process). We chose this example because it delineates all the essential points of our procedure (details of other N-photon processes will appear in another paper). In Fig. 1, the irreducible diagrams of the perturbation series for a four-photon process are drawn up to tenth order in the electromagnetic field. The vertical

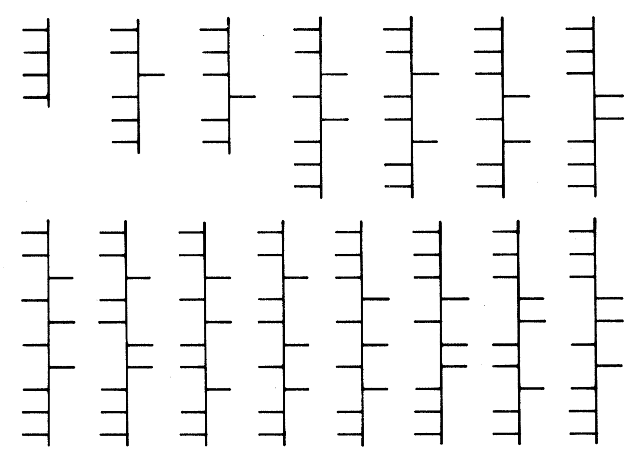

FIG. 1. The irreducible diagrams for a four-photon process up to tenth order in the electromagnetic field.

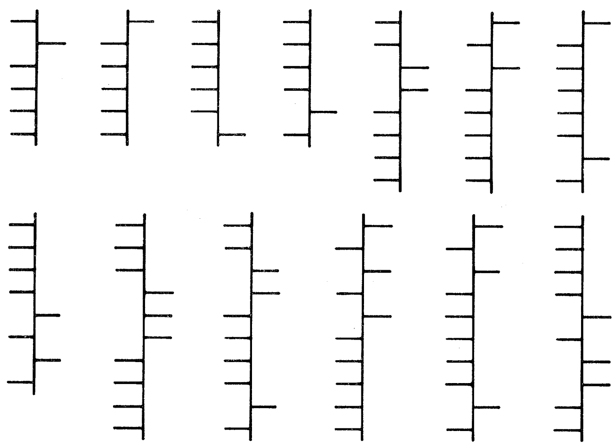

FIG. 2. Some of the reducible diagrams of the four-photon process up to tenth order.

line represents the electron in the potential of the atom; the horizontal lines are the photons emitted (to the right of the electron line) or absorbed (to the left of the electron line). In Fig. 2 some of the reducible diagrams are drawn. It is easy to see that any reducible diagram can be cut in a way such that a part is identical to one of the irreducible diagrams. The remaining part of the diagram consists of either the initial or the final state's emitting and absorbing equal numbers of photons. One can examine the analytic properties of the reducible diagrams and realize that each of the reducible diagrams is divergent, a situation common in the development of the perturbation series. We eliminate these divergences by renormalizing the initial and the final states of the atom. The remaining and the crucial task is then to sum the class of irreducible diagrams.

Consider the three elements of a diagram shown in Fig. 3. We represent Fig. 3(a) by a matrix element S_{ij}, Fig. 3(b) by another matrix element T_{ij}, and Fig. 3(c) by an element P_{ij}. The indices represent the states of the atom and each of these elements is constructed after summing over the intermediate states. The transition amplitude for the lowest order is given by $\sum_j S_{gj}P_{jf}$, where $|g\rangle$ and $|f\rangle$ represent the initial and the final states of the atom. The amplitude for the next higher order is given by $\sum_{jk} S_{gj}T_{jk}P_{kf}$. The entire series of the irreducible diagrams can be written down as

$$\mathcal{G}_{gf}^{(4)} = (SP)_{gf} + (STP)_{gf} + (ST^2P)_{gf} + \dots, \tag{1}$$

where $\mathcal{G}_{gf}^{(4)}$ is the transition amplitude for four-photon absorption. The series can be summed easily to produce

$$\mathcal{G}_{gf}^{(4)} = [S(1-T)^{-1}P]_{gf}. \tag{2}$$

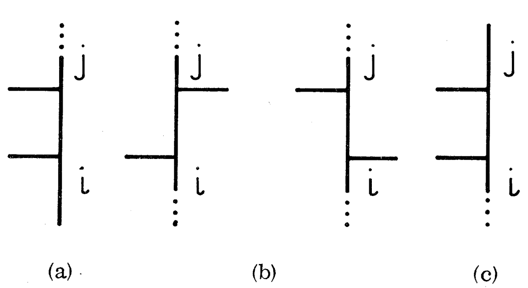

FIG. 3. The elements of the diagrams for a four-photon process.

The procedure for computation is straightforward. One constructs the matrix $1-T$ out of the element T_{ij}. The matrix is then inverted and finally one calculates, knowing the elements S_{gi} and P_{if}, the amplitude

$$\mathcal{G}_{gf}^{(4)} = \sum_{i,j} S_{gi}\left(\frac{1}{1-T}\right)_{ij} P_{jf}, \tag{3}$$

by summing over the states i and j. The dimension of the matrices is determined by the number of states chosen for computation. In Fig. 4, we have displayed the cross section for four-photon ionization of the ground state of the hydrogen atom as a function of the intensity of the external field. We have taken 28 states of the atom. This ensures sufficient stability of the result. The wavelength of the radiation is 3471.5 Å, which corresponds to the second harmonic of the ruby laser. At relatively low intensities, the cross section varies as F^3, where F is the photon flux (for N-photon absorption, the cross section varies as F^{N-1} for the lowest-order term of the perturbation series). As the intensity increases, the cross section departs from the behavior arising out of the lowest-order term of the perturbation series. There is a certain amount of structure caused by the higher-order terms. At 2.9×10^{14} W/cm^2, the limit of validity of the perturbation series is reached. Beyond this intensity, two sharp peaks are observed. These are flux-induced resonances[2] caused by the presence of the $2P$ and the $3P$ levels, respectively. It is interesting to note that in the nonperturbative region, the level structure of the atom continues to play an important role. With further increase of the incident photon flux, the cross section be-

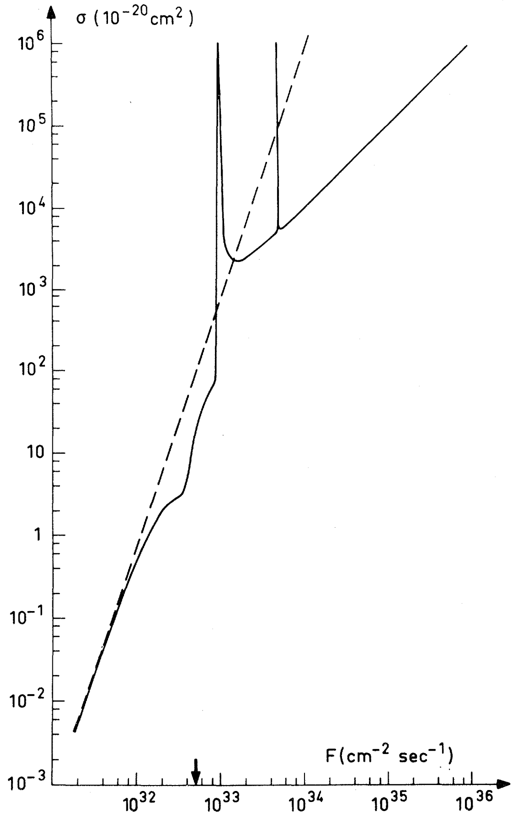

FIG. 4. Four-photon ionization cross section of H in the ground state as a function of the photon flux at 3471.5 Å (solid line). The dashed line represents the cross section from the lowest-order perturbation theory. The arrow indicates the limit of validity of the perturbation series.

haves smoothly and varies as F, which is remarkably different from the behavior at low intensities.

Our method allows us to vary the intensity continuously and to examine the behavior of the cross section over the entire region of intensities. Even at relatively low intensities, the contributions of the higher-order terms are taken into account. As the intensity increases, the contribution of the higher-order terms becomes more and more significant. At a certain intensity (depending on the atom as well as the photon number N), the perturbation series fails and this allows us to identify the limit of the validity of the perturbation series. Further, since we have closed-form expressions, we can examine the behavior of the cross section at arbitrarily large values of the photon flux. Here, the series given in Eq. (1) has no meaning, while the transition amplitude given by Eq. (2) continues to be valid as an analytic continuation to the nonperturbative region and thus transcends the usual perturbation theory.

In conclusion, two remarks are to be made. First, the method has been applied to higher N. The results (to be published in a complete paper) are more complicated because the irreducible diagrams are harder to sum. (There are more of them and they do not follow such simple recurrence relations as the example treated here.) We have succeeded in developing the algorithm to sum the series systematically. Second, the perturbation methods[2,3] proposed before do not sum the entire class of the irreducible diagrams, while the manifestly nonperturbative methods[4-7] proposed before lack the criteria to establish their connection with the entire perturbation series. Our proposed method takes into account the perturbation series exactly and can be analytically continued to the nonperturbative region.

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