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$E1$-$M1$ Interference in Radiative Decay of Hydrogenlike Atoms in an Electric Field

Peter J. Mohr
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720
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An unpolarized hydrogenlike atom in the metastable $2S_{1/2}$ state in an electric field decays primarily by one- or two-photon emission. The angular distribution of the single-photon radiation is expected to be asymmetric with respect to the electric field direction as a result of $E1$-$M1$ interference. Observable consequences of this effect in high-Z Lamb-shift experiments in progress are pointed out. A lowest-order estimate is given for the dependence of the asymmetry on the nuclear charge and the applied field strength.

Radiative decay of the $2S_{1/2}$ state in a static electric field has been studied in recent years as a means of determining the Lamb shift in hydrogen and hydrogenlike atoms. Both the lifetime of the excited state and the angular distribution of the emitted radiation, relative to the electric field direction, have been measured in order to infer values for the $2P_{1/2}$-$2S_{1/2}$ energy splitting.[1-3]

It is known that the angular distribution of electric-field–induced radiation from unpolarized hydrogenlike atoms is not isotropic, mainly because of the interference term between photon emission from the $2P_{1/2}$ and $2P_{3/2}$ states, which are mixed with the $2S_{1/2}$ state by the electric field. The anisotropy is characterized by a symmetric angular distribution of radiation of the form $a+b|\hat{k}\cdot\vec{E}|^2$, where $\vec{E}$ is the electric field vector and $\hat{k}$ is the direction of observation of the radiation. In high-Z hydrogenlike atoms, one may expect an additional asymmetric contribution (proportional to $\hat{k}\cdot\vec{E}$) to the angular distribution, which arises from interference between electric-field–induced electric dipole radiation and forbidden magnetic dipole radiation. For suitable combinations of nuclear charge and electric field strength, the ratio of the probability of photon emission in the direction of the electric field to the probability of emission in the opposite direction is less than 75%.

The purpose of this Letter is to point out the relevance of the asymmetry to high-Z Lamb-shift experiments now in progress, and give a lowest-order estimate of its magnitude. The asymmetry can be expected to play a role in the analysis of experiments based on measurement of the anisotropy of electric-field–induced radiation from the $2S_{1/2}$ state.[1] In these experiments, the relative intensity of radiation in the directions parallel and perpendicular to the field direction is measured, and a value for the Lamb shift is inferred from the ratio. The asymmetric term gives field-strength–dependent contributions of opposite sign to the intensity in the directions parallel and antiparallel to the electric field. The asymmetry should also be taken into account in experiments based on measurement of the lifetime of the $2S_{1/2}$ state in an electric field by the time-of-flight method.[2] Here, because of magnetic field bending of the metastable atomic beam, single-photon decays are viewed at a varying angle with respect to the electric field direction, and the observed intensity is affected by the asymmetric as well as the symmetric contributions to the angular distribution. Under the conditions of either of these experiments, the asymmetry is a measurable effect.

One can readily verify that an asymmetric term in the transition rate is consistent with time-reversal invariance, even though $\hat{k}$ changes sign under time reversal. An analogous situation arises, for example, in β decay where final-state Coulomb interactions give rise to terms which are proportional to combinations of polarization and momentum vectors, associated with the intitial and final states, which are odd under time reversal while the interaction is assumed to be time-reversal invariant.[4] In the case considered here, the decay rate is proportional to $a+b|\hat{k}\cdot\vec{E}|^2+c\Gamma\hat{k}\cdot\vec{E}$, where Γ is approximately equal to the radiative level width of the $2P_{1/2}$ state. The sign of Γ depends on the boundary conditions which determine the location of the pole in the electron propagation function. Inspection of the relevant Feynman integrals shows that the photon absorption rate is proportional to $a+b|\hat{k}\cdot\vec{E}|^2-c\Gamma\hat{k}\cdot\vec{E}$. Hence, the rates for the time-reversed processes, emission of a photon of momentum $\vec{k}$, and absorption of a photon of momentum $-\vec{k}$, are equal apart from overall kinematical factors.

In the absence of an external field, a hydrogenlike atom in the $2S_{1/2}$ state will radiatively decay to the $1S_{1/2}$ state primarily by either a two-photon transition or a one-photon forbidden magnetic dipole ($M1$) transition. For a nuclear charge Z, the two-photon transition rate is $A_{2\gamma}\approx 8.2Z^6$ sec^{-1} and the magnetic-dipole transition rate is $A_{M1}\approx 2.5\times10^{-6}Z^{10}$ sec^{-1}. At low Z, two-photon decay is dominant; however, because of the more rapid Z dependence of A_{M1}, magnetic dipole decay is more probable for $Z>40$.[5] In an applied electric field, one-photon electric dipole ($E1$) decays also occur with a transition rate given approximately by $A_{E1}{}^E\approx(3.1\times10^3)(E/S)^2Z^2$ sec^{-1}, where E (in units of V/cm) is the magnitude of the electric field and $S=E(2S_{1/2})-E(2P_{1/2})$ is the Lamb shift (in GHz) which scales roughly as Z^4. From the scaling of the rates, it is clear that the condition $A_{M1}\approx A_{E1}{}^E$, which maximizes the $E1$-$M1$ interference, is achieved with realistic electric fields over a range of Z for which the branching ratio for one-photon decays is not extremely small. For example, at $Z=18$, the condition $A_{M1}\approx A_{E1}{}^E$ occurs in an applied electric field $E=10^5$ V/cm, which is the motional electric field seen in the rest frame of a beam of atoms with velocity $v=0.1c$ passing through a laboratory magnetic field of 3.3 kG. In this case, 6% of the metastable atoms radiatively decay by one-photon emission.

The following simple estimate gives the magnitude and field dependence of the asymmetry. A more accurate calculation of the one-photon angular distribution and decay rate (which does not assume that Z is necessarily small) with a derivation based on the bound interaction formulation of quantum electrodynamics will be discussed in a separate paper in preparation. We consider here a hydrogenlike atom in a uniform, constant, and weak electric field $\vec{E}$, where weak means that the $2S_{1/2}$-$2P_{1/2}$ matrix element of the electric field perturbation is small compared to the Lamb shift, e.g., when $E(\mathrm{V/cm})\ll 10^2Z^5$. The effect of the electric field on the $2S_{1/2}$ state can be taken into account by applying first-order perturbation theory with the complex radiative level shifts included in the energy denominator. Only mixing of the $2S_{1/2}$ state with the $2P_{1/2}$ state is considered. We then have

$$|\overline{2S}_{1/2},\mu\rangle=|2S_{1/2},\mu\rangle+\eta\sum_{\mu''}\langle 2P_{1/2},\mu''|e\vec{E}\cdot\vec{x}|2S_{1/2},\mu\rangle|2P_{1/2},\mu''\rangle\,, \tag{1}$$

where

$$\eta=(S+i\Gamma_p/2)^{-1}, \tag{2}$$

and where μ,μ'' are the z components of the angular momentum, S is the Lamb shift, and Γ_p is the $2P_{1/2}$-state level width. The $2S_{1/2}$ width is negligible compared to Γ_p. The transition rate for $2S_{1/2}\rightarrow 1S_{1/2}$ with a photon emitted in the direction $\hat{k}$ (averaged over initial spins, summed over final spins, and summed over photon polarizations λ) is

$$\frac{dR}{d\Omega}=\frac{\alpha k}{2\pi}\frac{1}{2}\sum_{\mu\mu'\lambda}|M|^2\,, \tag{3}$$

(in units in which $\hbar=c=m_e=1$), where

$$M=\langle 1S_{1/2},\mu'|\hat{\epsilon}_\lambda\cdot\vec{\alpha}e^{-i\vec{k}\cdot\vec{x}}|\overline{2S}_{1/2},\mu\rangle\,. \tag{4}$$

In (3) and (4), $k=E(2S_{1/2})-E(1S_{1/2})$ and $\hat{\epsilon}_\lambda$ is the polarization vector of the photon. Substitution of (1) into (4), keeping only the leading terms in $(Z\alpha)^2$, yields

$$M=-(81\sqrt{2})^{-1}\langle\mu'|3(Z\alpha)^4\vec{\sigma}\cdot\hat{\epsilon}_\lambda\vec{\sigma}\cdot\hat{k}-32i\eta\vec{\sigma}\cdot\hat{\epsilon}_\lambda\vec{\sigma}\cdot\vec{F}|\mu\rangle\,, \tag{5}$$

where $|\mu\rangle$ is a two-component vector with upper and lower components $\frac{1}{2}+\mu$ and $\frac{1}{2}-\mu$, with μ and μ' taking on the values $\pm\frac{1}{2}$, and where $\vec{F}=e\vec{E}$. The first term in (5) is the forbidden magnetic dipole decay contribution and the second term is the electric-field–induced electric-dipole decay contribution.

From (3) and (5), the differential decay rate is

$$\frac{dR}{d\Omega}=\frac{\alpha k}{13122\pi}\left(9(Z\alpha)^8+\frac{1024F^2}{S^2+\Gamma_p^2/4}-\frac{96(Z\alpha)^4\Gamma_p}{S^2+\Gamma_p^2/4}\hat{k}\cdot\vec{F}\right). \tag{6}$$

The cross term gives a field-dependent asymmetry proportional to Γ_p.

An asymmetry parameter A may be defined in terms of the rate for photon emission in the direction of the electric field I_+ and the rate for emission in the opposite direction I_- as $A=(I_+-I_-)/(I_++I_-)$:

$$A=-\frac{96(Z\alpha)^4\Gamma_p F}{9(Z\alpha)^8(S^2+\Gamma_p^2/4)+1024F^2}. \tag{7}$$

The maximum magnitude of A, with respect to the variation of F, is

$$A_{\max}=-\tfrac{1}{2}\Gamma_p/(S^2+\tfrac{1}{4}\Gamma_p^2)^{1/2}, \tag{8}$$

which occurs when the two terms in the denominator of (7) are equal, i.e., when $A_{M1}=A_{E1}{}^E$. The maximum asymmetry is a slowly varying function of Z with the values $A_{\max}=-9\%$ at $Z=8$ and $A_{\max}=-14\%$ at $Z=18$. For the special case $A_{M1}\ll A_{E1}{}^E$, which pertains to the experiments of Drake[1] and of Gould and Marrus,[2] A depends only weakly on S, and is given approximately by

$$A\approx-\frac{3}{32}(Z\alpha)^4\frac{\Gamma_p}{F}=-\frac{(2.8\times10^{-6})Z^8}{E} \tag{9}$$

to lowest order in $(Z\alpha)^2$, where E is in units of V/cm.

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Self-Consistent Relativistic Molecular Calculations of Superheavy Molecules: $({}_{110}X)F_6$

A. Rosén
Chalmers University of Technology, Göteborg, Sweden

and

B. Fricke and T. Morović
Gesamthochschule Kassel, D-3500 Kassel, West Germany
(Received 28 October 1977)

Using new relativistic molecular calculations within the Dirac-Slater scheme it is now feasible to study theoretically molecules containing superheavy elements. This opens a new era for the prediction of the physics and chemistry of superheavy elements. As an example we present the results for $({}_{110}X)F_6$, where it is shown that relativistic effects are nearly of the same order of magnitude as the crystal-field splitting.

In recent years a number of atomic calculations have been carried out in the region of superheavy elements. With sophisticated interpretations of these results one was able to perform relatively realistic extrapolations and thus got a first idea of the physical and chemical properties of the still-unknown superheavy elements. These results have been reviewed by Hermann[1] and Fricke.[2] Many authors[1-3] also have emphasized that further development in this field is dependent

