Calculation of Stark-induced absorption on the $6s6p\ ^3P_1-6s^2\ ^1S_0$ transition in Hg

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We carry out relativistic many-body calculations of the Stark-induced absorption coefficient on the 254-nm $6s6p\ ^3P_1(F=1/2)-6s^2\ ^1S_0$ line of ^{199}Hg atom, the effect considered before by and Lamoreaux and Fortson [Phys. Rev. A **46**, 7053 (1992)] using a simple central field estimate. The Stark-induced admixing of states of opposite parity opens additional $M1$ and $E2$ transition channels. We find that the resulting $M1$-$E1$ absorption dominates over $E2$-$E1$ absorption. The value of the $E2$-$E1$ absorption coefficient depends strongly on the details of treatment of the correlation problem. As a result, our numerical values differ substantially from those of the earlier central field calculation. Reliable calculation of this effect can enable a useful experimental check on the optical technique being used to search for a permanent electric-dipole moment of the ^{199}Hg atom.

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I. INTRODUCTION

The $F=1/2\rightarrow F=1/2$ electromagnetic transition between two atomic states of opposite parity has necessarily the electric-dipole ($E1$) character. However, an application of the external electric field $\mathcal{E}_s$ breaks the spherical and mirror symmetries of the atomic Hamiltonian and opens all multipolar transition channels. To the lowest order in $\mathcal{E}_s$, the transitions are determined by the $M1$ (magnetic-dipole) and $E2$ (electric-quadrupole) channels. These effects modify the absorption coefficient of the atomic sample, the corrective $M1$ and $E2$ terms scaling linearly with the electric field [1,2].

Lamoreaux and Fortson [2] focused on a specific setup relevant for the search of the permanent electric-dipole moment (EDM) of Hg atom [3,4] (nonvanishing EDM would violate P- and T-reversal symmetries and be a clear signature of new physics beyond the standard model of elementary particles). They considered exciting the 254-nm $6s^2\ ^1S_0\rightarrow 6s6p\ ^3P_1$ transition of ^{199}Hg atom. This isotope has the nuclear spin $I=1/2$. A laser resolves the hyperfine structure of the $^3P_1$ level. It resonantly drives transitions from a given magnetic M_{F_i} sublevel of the $F_i=1/2$ ground state to the $F_f=1/2$ level of the excited state. Then, for the $F_i=1/2\rightarrow F_f=1/2$ transitions, the relative change in the absorption coefficient α may be parameterized as [2]

$$\frac{\delta\alpha}{\alpha}=(a_{M1}+a_{E2})(\hat{\varepsilon}_L\cdot\mathcal{E}_s)(\hat{\mathbf{k}}_L\times\hat{\varepsilon}_L)\cdot\left(\frac{\langle\mathbf{F}_i\rangle}{F_i}\right). \tag{1}$$

Here $\hat{\varepsilon}_L$ is the polarization vector and $\hat{\mathbf{k}}_L$ is the direction of propagation of the laser wave. $\langle\mathbf{F}_i\rangle$ is the expectation value of the total angular momentum, i.e., the nuclear polarization in the ground $J_i=0$ atomic state.

In the current ^{199}Hg EDM experiment [4], the 254-nm transition is used to monitor the nuclear-spin direction and thereby detect EDM-induced shifts in nuclear-spin precession, which will be linear in an external electric field. The Stark interference effect on the 254 nm absorption given in Eq. (1) also is linear in electric field $\mathcal{E}_s$ and depends upon the nuclear-spin direction. A reliable calculation of this effect can enable a useful check on the EDM method when the effect is measured under the same experimental conditions as in the EDM search [5].

II. EXPRESSIONS FOR ABSORPTION COEFFICIENTS

The goal of this paper is to compute the atomic-structure coefficients a_{M1} and a_{E2}. One may qualitatively understand the appearance of $M1$ and $E2$ admixtures in Eq. (1) as follows. The Stark-induced transition amplitude in a laser field is composed from terms linear in the interactions with the external electric field, $-\mathbf{D}\cdot\mathcal{E}_s$ ($\mathbf{D}$ being the dipole operator), and the driving 2^k-pole laser field. We may recouple the products of the two tensors ($\mathbf{D}$ and the laser EM multipolar interaction); the resulting compound operators have the multipolarities of $|k-1|,k,k+1$. For the $F_i=1/2\rightarrow F_f=1/2$ transition, k would be limited to 1 and 2. The additional constraint imposed by the parity selection rule yields the $M1$ and $E2$ multipolar couplings.

We derived the expressions for a_{M1} and a_{E2} using the multipolar expansion of the plane EM wave and the first-order perturbation theory in the Stark field for the wave function. We employ a geometry where the quantization axis $\hat{z}$ is chosen along the k vector of the linearly polarized laser. The dc Stark field and the laser polarization are aligned along the x axis, and the atom has a definite value of $\hat{F}_y$ in the initial state. This particular choice of geometry is convenient for working with the most general relativistic expressions for the multipolar transition operators [6]. By evaluating Eq. (1) of Ref. [2] in this geometry, we identify the following expressions for the structure factors:

$$a_{M1}=\sqrt{\frac{2}{3}}\frac{R^{M1}}{\langle n_iJ_i||\mathbf{r}||n_fJ_f\rangle}, \tag{2}$$

$$a_{E2}=-\frac{1}{4}\sqrt{\frac{3}{5}}k_L\frac{R^{E2}}{\langle n_iJ_i||\mathbf{r}||n_fJ_f\rangle}. \tag{3}$$

Unless specified otherwise, here and below we use the atomic units, $\hbar=|e|=m_e=1$, and the Gaussian units for EM

equations. Here $\langle n_i J_i||\mathbf{r}||n_f J_f\rangle$ is (within a factor of -1 corresponding to the charge of the electron) the conventional reduced dipole matrix element for the $6s^2\ {}^1S_0 \rightarrow 6s6p\ {}^3P_1$ transition and $k_L=\omega_L/c$ is the magnitude of the wave vector of the laser. The quantities R^{M1} and R^{E2} are sums over a complete set of intermediate states; these sums arise due to the Stark-induced perturbation and involve the static $E1$ operator and multipolar ac couplings to the driving laser field. Each of the sums, R^{M1} and R^{E2}, may be further split into two sums $S_{i/f}^{M1/E2}$, subscript i or f indicating which of the states, initial or final, is Stark-perturbed,

$$R^{M1} = -S_i^{M1}(1^o) + S_f^{M1}(1^e), \tag{4}$$

$$R^{E2} = -\tfrac{2}{3}\sqrt{2}S_i^{E2}(1^o) - 2\sqrt{\tfrac{2}{15}}S_f^{E2}(2^e). \tag{5}$$

The argument of the sums $S_{i/f}^{M1/E2}(J^\pi)$ indicates the total angular momentum J and the parity π of the intermediate states as fixed by selection rules. Explicitly, the reduced sums are

$$S_i^{M1}(J_n^\pi = 1^o) = \sum_n \frac{\langle n_i J_i||\mathbf{r}||n_n J_n\rangle\langle n_n J_n||Q^{(M1)}||n_f J_f\rangle}{E_i - E_n},$$

$$S_f^{M1}(J_n^\pi = 1^e) = \sum_n \frac{\langle n_i J_i||Q^{(M1)}||n_n J_n\rangle\langle n_n J_n||\mathbf{r}||n_f J_f\rangle}{E_f - E_n},$$

$$S_i^{E2}(J_n^\pi = 1^o) = \sum_n \frac{\langle n_i J_i||\mathbf{r}||n_n J_n\rangle\langle n_n J_n||Q^{(E2)}||n_f J_f\rangle}{E_i - E_n},$$

$$S_f^{E2}(J_n^\pi = 2^e) = \sum_n \frac{\langle n_i J_i||Q^{(E2)}||n_n J_n\rangle\langle n_n J_n||\mathbf{r}||n_f J_f\rangle}{E_f - E_n}.$$

We employ the relativistic formalism for the multipolar transition operators $Q^{(M1/E2)}$. Specific single-particle reduced matrix elements computed using Dirac orbital parameterization of Ref. [6] are

$$\langle i||Q^{(EJ)}||j\rangle = \langle \kappa_i||C_J||\kappa_j\rangle \int_0^\infty r^J\{G_i(r)G_j(r) + F_i(r)F_j(r)\}dr, \tag{6}$$

$$\langle i||Q^{(MJ)}||j\rangle = \frac{\kappa_i + \kappa_j}{J+1}\langle -\kappa_i||C_J||\kappa_j\rangle \times \int_0^\infty r^J\{G_i(r)F_j(r) + F_i(r)G_j(r)\}dr. \tag{7}$$

In both expressions we used the long-wavelength approximation, as $\alpha\omega_L \ll 1$. In these expressions, $G(r)$ $[F(r)]$ are the large (small) radial components of the Dirac bi-spinor, κ are the relativistic angular quantum numbers, and $C_J(\hat{r})$ are the normalized spherical harmonics.

III. ATOMIC-STRUCTURE FORMALISM

Mercury atom has two valence electrons outside a closed-shell core and we start our calculations with the so-called frozen core (V^{N-2}) Dirac-Hartree-Fock (DHF) approximation. In this approximation, the core orbitals are obtained self-consistently, while excited (valence) orbitals are subsequently generated by solving the Dirac equation in the resulting potential of the core. Such orbitals correspond to the Hg^+ valence orbitals. They are used as a basis for the standard configuration-interaction (CI) technique for two valence electrons (see, e.g., [7].). We refer to this approximation as CI-DHF. Further significant improvement of the accuracy of the calculations is achieved when the standard CI technique is combined with many-body perturbation theory (MBPT) to include correlations of the valence electrons with the atomic core (CI+MBPT).

The CI+MBPT formalism has been discussed in a number of papers (see, e.g., [8–10]). The effective operator (self-energy $\hat{\Sigma}$) arising from the core polarization may be split into a single-particle, $\hat{\Sigma}_1$, and a two-particle, $\hat{\Sigma}_2$, part acting in the model space. Qualitatively, a field of the valence electron induces an electric dipole of the polarizable core: $\hat{\Sigma}_1$ describes an interaction of the valence electron with the self-induced core dipole, while $\hat{\Sigma}_2$ describes its interaction with the core dipole induced by the *other* valence electron. We compute the self-energy correction in the second order of MBPT for the residual Coulomb interaction. Effects of higher orders will be also included in a semiempirical fashion, discussed below.

We use the Brillouin-Wigner flavor of MBPT [7] to avoid the "intruder-state problem" when the virtual core excitations inside $\hat{\Sigma}_2$ become resonant with the states of the valence subspace. Finally, we emphasize that our computations are *ab initio* relativistic and employ the Dirac equation and bi-spinors throughout the entire calculation.

We use the second-order MBPT to calculate the self-energy operators $\hat{\Sigma}_1$ and $\hat{\Sigma}_2$ via direct summation over a complete set of single-electron states. This set of basis states is constructed using the B-spline technique [11]. We use 40 B splines of order 9 in a cavity of 40 Bohr radius. The same basis of the single-electron states is also used in constructing the two-electron basis states for the CI calculations. We employ partial waves $\ell=0-4$ and the 14 lowest states above the core in each partial wave ($s_{1/2}$, $p_{1/2}$, $p_{3/2}$, etc.) for the valence CI subspace and $\ell=0-5$ and 30 lowest states in each partial wave for internal summations inside the self-energy operator.

Higher-order correlations are also included in $\hat{\Sigma}$ in a way similar to Ref. [12]. The $\hat{\Sigma}_1$ operator depends on the symmetry of the valence orbital. Therefore, we have a set of different $\hat{\Sigma}_1$ operators for $s_{1/2}$, $p_{1/2}$, $p_{3/2}$, etc., states. An analysis of the spectra of Hg (see Table I) shows that accurate treatment of $\hat{\Sigma}_1$ is most important for s electrons because the ground $6s^2$ state and other states with s electrons come close to the core and therefore core-valence correlations must be sizeable for them. In contrast, the core-valence correlations are much smaller for more diffuse p and d orbitals. It turns out that the best accuracy is achieved if the all-order $\hat{\Sigma}_1^\infty$ [13] operator is used for the s electrons, the second-order $\hat{\Sigma}_1$ is employed for the p electrons, and no $\hat{\Sigma}_1$ is included for d and higher waves.

TABLE I. Experimental and theoretical energy levels of Hg (in cm^{-1}).

State		J	Expt. [14]	Theory
$6s^2$	^1S	0	0.000	−13.79
$6s6p$	$^3P^o$	0	37645.080	37458.26
		1	39412.300	39312.86
		2	44042.977	44265.45
$6s6p$	$^1P^o$	1	54068.781	54180.72
$6s7s$	^3S	1	62350.456	62171.92
$6s7s$	^1S	0	63928.243	63672.24
$6s7p$	$^3P^o$	0	69516.66	69211.87
		1	69661.89	69385.18
		2	71207.51	70094.95
$6s7p$	$^1P^o$	1	71295.15	71189.34
$6s6d$	^1D	2	71333.182	71295.01
$6s6d$	^3D	2	71396.220	71353.26

Higher-order contributions to $\hat{\Sigma}_2$ are included semiempirically via screening factors which modify Coulomb integrals of the second-order $\hat{\Sigma}_2$ (see Ref. [12] for details). The values of these factors are $f_0=0.9$, $f_1=0.72$, $f_2=0.98$, $f_3=1$, $f_4=1.02$, and $f_5=1.02$. These values are found from comparing second-order and all-order $\hat{\Sigma}_1$.

Finally, we further rescale the $\hat{\Sigma}_1$ operator for the s and p electrons to fit the experimental spectrum better. The rescaling coefficients are $\lambda_s=1.0961$ and $\lambda_p=0.8675$. We use the same λ_p for $p_{1/2}$ and $p_{3/2}$ waves. Note that $\lambda_s>1$ because high-order effects, included in $\hat{\Sigma}_1$ for the s electrons, significantly reduce its value. On the other hand, $\lambda_p<1$ because the second-order MBPT always overestimates the correlation correction.

The resulting energies are listed and compared with experiment in Table I. A typical deviation from the experimental values is in the order of 100 cm^{-1}. Even after the scaling, the disagreement remains, as the number of fitting parameters is limited.

The diagonalization of the CI+MBPT Hamiltonian provides us with the atomic wave functions and energies. While the wave functions already have correlation corrections built in, evaluating matrix elements requires additional inclusion of the so-called screening effect. This effect arises already in the first order in the residual Coulomb interaction and describes a re-adjustment of the core orbitals in response to an externally applied field. We incorporate the screening in the framework of the all-order many-body technique, the random-phase approximation (RPA). The RPA formalism (see, e.g., Ref. [15]) describes a linearized response to an oscillating perturbation. In this regard, while evaluating the reduced sums, we need to fix the driving RPA frequency for the entire set of matrix elements $Q^{(M1)}$ and $Q^{(E2)}$ at the photon frequency, $\omega_L=E_f-E_i$. However, for the dipole matrix elements (Stark mixing), the RPA frequency $\omega=0$.

TABLE II. Energy interval ΔE (in cm^{-1}) and the reduced electric-dipole matrix element (R,a.u.) for the $6s^2\ ^1S_0-6s6p\ ^3P_1$ transition in Hg atom in various approximations.

Approximation	ΔE	R
CI-DHF	31028	0.405
CI+Σ_1	37441	0.453
CI+Σ_1+Σ_2	37623	0.716
CI+Σ_1+Σ_2+RPA		0.577
As above but with all-order Σ_2	36947	0.512
As above but with scaled Σ_1 (Final)	39313	0.503
Expt. [14,18]	39412	0.453(8)

The evaluation of the sums S requires summing over a complete set of intermediate atomic states $|n_nJ_n\rangle$. We use two approaches: (i) direct summation over states (this implies explicit computation of the atomic states and evaluation of matrix elements) and (ii) the Dalgarno-Lewis method. In the Dalgarno-Lewis method [16], the summation is reduced to solving the inhomogeneous Schrödinger (Dirac) equation (setup is similar to Ref. [17]). As an illustration, consider evaluation of the sum S_f^{M1}. It may be represented as

$$S_f^{M1}=\langle n_iJ_i||Q^{(M1)}||\delta\Psi_f\rangle,$$

where $|\delta\Psi_f\rangle$ lumps corrections to the atomic wave function of the final state due to the external field. It satisfies an inhomogeneous equation

$$(\hat{H}_{\rm eff}-E_f)|\delta\Psi_f\rangle=-\mathbf{r}|n_fJ_f\rangle, \tag{8}$$

where $\hat{H}_{\rm eff}$ is the effective CI+MBPT Hamiltonian of the atom.

IV. NUMERICAL RESULTS

As an illustration of the CI+MBPT methodology, we start with calculations of the $E1$ matrix element and energy interval for the $6s^2\ ^1S_0-6s6p\ ^3P_1$ transition. This matrix element normalizes the Stark-induced corrections to the absorption coefficient, Eq. (1). The theoretical results at various levels of approximation and a comparison with the experimental values are presented in Table II. We observe that the core polarization ($\hat{\Sigma}_1$) has a substantial effect on the energy interval, leading to an improvement in the theory-experiment agreement. While the CI+$\hat{\Sigma}_1$ value of the matrix element perfectly agrees with the experiment [18], such an agreement is fortuitous: including the screening correction to the Hamiltonian ($\hat{\Sigma}_2$) increases its value by a factor of 1.6; only the additional inclusion of the RPA screening and semiempirical scaling moves the theoretical value into a 10% agreement with a 2%-accurate experiment. We find such an accuracy acceptable as *ab initio* matrix elements of the intercombination (spin-forbidden) transitions are known [19] to be very sensitive to many-body corrections, the entire values being accumulated due to the relativistic effects. On the other hand, the matrix elements of *spin-allowed* transitions are stable

TABLE III. Breakdown of contributions to the reduced sums for the $E2$ Stark-induced transition. All quantities are in atomic units.

	$S_i^{E2}(J_n^\pi=1^o)$		
n_nJ_n	$\langle 6s^2\ ^1S_0\|\|\mathbf{r}\|\|n_nJ_n\rangle$	$\langle n_nJ_n\|\|Q^{(E2)}\|\|6s6p\ ^3P_1\rangle$	Contribution
$6s6p\ ^3P_1$	−0.503	7.949	22.29
$6s6p\ ^1P_1$	−2.956	−4.535	−54.41
$6s7p\ ^3P_1$	−0.037	−5.460	−0.63
$6s7p\ ^1P_1$	0.674	−1.647	3.42
$6s8p\ ^3P_1$	−0.005	1.839	0.03
$6s8p\ ^1P_1$	−0.286	1.652	1.35
$6s9p\ ^3P_1$	−0.063	−2.875	−0.50
$6s9p\ ^1P_1$	0.314	−0.269	0.23
Sum(10)			−28.23
Dalgarno-Lewis, Sum (∞)			−25.60
	$S_f^{E2}(J_n^\pi=2^e)$		
n_nJ_n	$\langle 6s6p\ ^3P_1\|\|\mathbf{r}\|\|n_nJ_n\rangle$	$\langle n_nJ_n\|\|Q^{(E2)}\|\|6s^2\ ^1S_0\rangle$	Contribution
$6s6d\ ^1D_2$	1.570	−6.963	75.18
$6s6d\ ^3D_2$	2.360	3.248	−52.61
$6s7d\ ^1D_2$	0.576	−6.153	20.67
$6s7d\ ^3D_2$	1.354	1.661	−13.10
$6s8d\ ^1D_2$	0.263	−2.857	4.10
$6s8d\ ^3D_2$	−1.363	0.422	3.14
$6s9d\ ^1D_2$	−0.284	3.654	5.47
$6s9d\ ^3D_2$	−1.281	−0.549	−3.71
Sum(10)			39.14
Dalgarno-Lewis, Sum (∞)			44.97

with respect to inclusion of the MBPT effects (see, e.g., Ref. [19]). We will return to the evaluation of the accuracy of our calculations later.

The Stark-induced correction to the absorption coefficient involves two channels, $M1$ and $E2$. We start by discussing the more involved a_{E2} calculations. We need to compute two sums, $S_i^{E2}(J_n^\pi=1^o)$ and $S_f^{E2}(J_n^\pi=2^e)$. We carry out calculations (i) by direct summation over the ten lowest-energy intermediate states of each symmetry (1^o and 2^e) and (ii) by using the Dalgarno-Lewis method. The latter method is equivalent to summing over infinitely many intermediate states. Both calculations use the most sophisticated CI+MBPT approximation (i.e., CI+Σ_1+Σ_2+RPA with semiempirical scaling). The results are presented in Table III. An examination of contributions reveals that there are substantial cancellations inside individual sums. This leads to an enhanced sensitivity to correlations. For example, consider the value of the $S_i^{E2}(J_n^\pi=1^o)$ sum truncated at the ten lowest-energy levels. It changes from −35.6 to −28.23 (Table III) while progressing from the CI+Σ_1+RPA to the full CI+MBPT treatment. Additional cancellations occur when the reduced sums are combined into the quantity $R^{E2}=-\frac{2}{3}\sqrt{2}S_i^{E2}(1^o)-2\sqrt{\frac{2}{15}}S_f^{E2}(2^e)\approx 24.13-32.84=-8.71$. Notice that this value is several times smaller than the properly rescaled value of the largest contribution in Table III. These cancellations may lead to a poor accuracy of our resulting absorption coefficient

$$a_{E2}=-4.39\times 10^{-3}\ \text{a.u.}=-0.0853\times 10^{-8}(\text{kV/cm})^{-1}.$$

This result was obtained using the *ab initio* matrix element from Table II. Notice that there is a phase ambiguity originating from atomic wave functions for sums S and the normalizing dipole matrix element. However, when these quantities are combined in Eqs. (2) and (3), the ambiguous phase factors cancel out. In our particular computation, the sign of the dipole matrix element $\langle 6s^2\ ^1S_0\|\mathbf{r}\|6s6p\ ^3P_1\rangle$ is fixed by the first entry of Table III.

We proceed to a comparison with results of Ref. [2]. These authors use a simplified approach in which a true many-electron problem is reduced to a set of single-electron problems. For the $E2$ interference they use the Dalgarno-Lewis summation method based on the DHF orbitals of the optically active valence electron. The LS coupling scheme was used in calculations. Their P (D)-channel results correspond to our 1^o (2^e) values. We find some numerical errors in calculations of the a_{E2} coefficient and so a comparison is hindered a bit. For example, for the P channel, using Eq. (39) of Ref. [2] and their numerical values we obtain, $a_{E2,P}=-0.96\times 10^{-8}$ cm/kV which is an order of magnitude

TABLE IV. Comparison of different multipolar contributions to the Stark-induced absorption coefficients a_{M1} and a_{E2} in 1/(kV/cm). The first column gives the character of the multipole, the second column lists values of Ref. [2], and the third column gives the results of our computation. The notation $a[b]=a\times 10^b$ is used.

Contribution	Ref. [2]	This work
$E2$, 1^o	−9.6[−9] [a]	2.4[−9]
$E2$, 2^e	1.0[−9] [a]	−3.2[−9]
$E2$, total	−8.7[−9] [a]	−0.85[−9]
$M1$, 1^e	0	−0.13[−9]
$M1$, 1^o	7.8[−9]	8.9[−9]
$M1$, total	7.8[−9]	8.86[−9]

[a]Values recomputed by us based on data of Ref. [2]; there are errors in numerical evaluations of Eqs. (40), (46), and (47) of Ref. [2]. See text for details.

smaller than the published value. Similarly for the D channel, based on Eq. (46) and numerical values of Ref. [2], we find $a_{E2,D}=0.1\times 10^{-8}$ cm/kV, a factor of 20 smaller than the published value. We present a detailed comparison with (revised values of) Ref. [2] for the two symmetries of intermediate states (1^o and 2^e) in Table IV. Although of the same order of magnitude, the individual contributions differ by signs. The most probable reason for the disagreement is the sensitivity to particulars of the treatment of correlations. For example, in computation of the D-channel contribution, Ref. [2] omitted intermediate states of the ^3D symmetry. Hg is a heavy atom, and according to Table III, omitting the triplet contributions would increase $a_{E2,D}$ by a factor of three. There is a remarkable cancellation between individual channels (Table IV): our final result becomes an order of magnitude smaller than the recomputed value ($a_{E2}=-0.87\times 10^{-8}$ cm/kV) of Ref. [2].

Fortunately, while a_{E2} has a poor accuracy, it turns out to be much smaller than a_{M1}, which, as shown below, can be computed reliably. There are two reduced sums to evaluate, $S_i^{M1}(1^o)$ and $S_f^{M1}(1^e)$. Nonrelativistically, the magnetic-dipole operator is diagonal in the radial quantum numbers. This means that the only substantial contributions arise in the sum $S_i^{M1}(1^o)$. Indeed, we find from our fully relativistic analysis

$$S_i^{M1}(1^o)\approx 0.0285,$$

$$S_f^{M1}(1^e)\approx 0.0004.$$

The two dominant matrix elements entering $S_i^{M1}(1^o)$ are $\langle 6s6p\ ^3P_1||Q^{(M1)}||6s6p\ ^3P_1\rangle$ and $\langle 6s6p\ ^3P_1||Q^{(M1)}||6s6p\ ^1P_1\rangle$. Both matrix elements may be estimated nonrelativistically (e.g., one could use the Landé formula for the first matrix element). Further, the term involving the $|6s6p\ ^3P_1\rangle$ state is larger by roughly a factor of 5 than the contribution from the singlet state. As a result, the uncertainty in evaluating $S_i^{M1}(1^o)$ comes from the dipole matrix element entering this contribution, the already discussed $\langle 6s6p\ ^3P_1||\mathbf{r}||6s^2\ ^1S_0\rangle$. Incidentally, this is the very same matrix element that normalizes the absorption coefficient so it cancels out in a_{M1}. Therefore, with about 25% accuracy

$$a_{M1}\approx\sqrt{\frac{2}{3}}\frac{\langle ^3P_1||Q^{(M1)}||^3P_1\rangle}{E_f-E_i}\approx 1.19\times 10^{-8}/(\mathrm{kV/cm}),$$

where we used the nonrelativistic value $\langle ^3P_1||Q^{(M1)}||^3P_1\rangle=(\frac{3}{4})\sqrt{6}\alpha$. Our full-scale Dalgarno-Lewis relativistic CI+MBPT calculation results in

$$a_{M1}=0.886\times 10^{-8}/(\mathrm{kV/cm}),$$

and is consistent with the nonrelativistic estimate. From the preceding discussion, it is clear that our theoretical value is stable with respect to neglected many-body corrections. Ref. [2] arrived at the result $a_{M1}=0.780\times 10^{-8}/(\mathrm{kV/cm})$. This differs by 12% from our calculations.

Finally, we combine the contributions of the $M1$ and $E2$ interferences. We note that the poorly known $E2$ contribution is fortunately suppressed by a factor of 10 compared to the $M1$ coefficient. We find

$$a_{M1}+a_{E2}=0.80\times 10^{-8}/(\mathrm{kV/cm}).$$

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