Reply to "Comment on 'Phenomenological damping in optical response tensors' "

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We show that damping factors must not be incorporated in the perturbation of the ground state by a static electric field. If they are included, as in the theory of Stedman *et al.* [preceding Comment, Phys. Rev. A **63**, 047801 (2001)], then there would be an electric dipole in the y direction induced in a hydrogen atom in the $M_S=+\frac{1}{2}$ state by a static electric field in the x direction. Such a dipole is excluded by symmetry.

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We note that Stedman *et al.* (SNAD) [1] now agree that optical susceptibilities in the polarization formalism should have the signs of damping factors as in Bloembergen [2,3], Flytzanis [4], Shen [5], Butcher and Cotter [6], and our paper [7]. An important issue remaining, however, is the question of whether damping is "inevitable from any perturbation, including a static field" [1]. Andrews *et al.* [8] and SNAD assert that it is inevitable. We shall address this important issue by considering the response of a spherical atom to a static electric field and shall show that the assertion is false.

Suppose that damping is necessary in the description of the first-order perturbed ground-state ket of an atom in a static electric field F_β. Such a ket may then be written in the form

$$|g_F^{\mathrm{damp}}\rangle=|g\rangle+\sum_{j\neq g}\frac{\langle j|\hat{\mu}_\beta|g\rangle}{\hbar\left(\omega_{jg}-\frac{\mathrm{i}}{2}\Gamma_{jg}\right)}|j\rangle F_\beta, \tag{1}$$

where $\hat{\boldsymbol{\mu}}$ is a dipole operator, ω_{jg} is the transition angular frequency between the upper level j and the ground state g, and Γ_{jg} is the associated damping factor. The summation is over all excited states $j\neq g$. The expectation value of the α component of the atom's dipole moment in the field would then be

$$\mu_\alpha=\langle g_F^{\mathrm{damp}}|\hat{\mu}_\alpha|g_F^{\mathrm{damp}}\rangle=\sum_{j\neq g}\left\{\frac{\langle g|\hat{\mu}_\alpha|j\rangle\langle j|\hat{\mu}_\beta|g\rangle}{\hbar\left(\omega_{jg}-\frac{\mathrm{i}}{2}\Gamma_{jg}\right)}+\frac{\langle g|\hat{\mu}_\beta|j\rangle\langle j|\hat{\mu}_\alpha|g\rangle}{\hbar\left(\omega_{jg}+\frac{\mathrm{i}}{2}\Gamma_{jg}\right)}\right\}F_\beta$$

$$=\sum_{j\neq g}\frac{2\omega_{jg}\,\mathrm{Re}\{\langle g|\hat{\mu}_\alpha|j\rangle\langle j|\hat{\mu}_\beta|g\rangle\}-\Gamma_{jg}\,\mathrm{Im}\{\langle g|\hat{\mu}_\alpha|j\rangle\langle j|\hat{\mu}_\beta|g\rangle\}}{\hbar\left(\omega_{jg}^2+\frac{1}{4}\Gamma_{jg}^2\right)}F_\beta. \tag{2}$$

If $|g\rangle$ is degenerate, as for a hydrogen or sodium atom in an $M_S=\pm\frac{1}{2}$ spin state, then $|g\rangle$ is complex, and both the real and imaginary parts of $\langle g|\hat{\mu}_\alpha|j\rangle\langle j|\hat{\mu}_\beta|g\rangle$ are nonzero. The imaginary part changes sign on interchanging α and β. This would imply that a static field F_x induces a dipole in the y direction in a spherical atom in an $M_S=+\frac{1}{2}$ state. However, this is incompatible with quantum theory, for such a dipole must be zero by symmetry (it would be equal and opposite for the $M_S=+\frac{1}{2}$ and $-\frac{1}{2}$ states and therefore change sign under time reversal).

If $|g\rangle$ is nondegenerate, as for a helium atom, then $|g\rangle$ may, without loss of generality, be chosen to be real. The induced dipole would be

$$\mu_x=\sum_{j\neq g}\frac{2\omega_{jg}|\langle g|\hat{\mu}_x|j\rangle|^2}{\hbar^2\left(\omega_{jg}^2+\frac{1}{4}\Gamma_{jg}^2\right)}F_x=\alpha F_x, \tag{3}$$

which is an incorrect equation for the static polarizability α. The correct equation for α is obtained from Eq. (3) by omitting all Γ_{jg}.

We note that SNAD appear to have missed the notes in the epilogue to Bloembergen's book [2]; the section SNAD refer to has been replaced by the following statement: "The correct limiting behavior for the case that either the electromagnetic frequency or the material resonant frequency becomes very small, $\omega\to 0$ or $\omega_{ng}\to 0$, respectively, requires a more careful treatment of the damping terms, as has been discussed in detail by Van Vleck and Weisskopf [9]."

SNAD do not refute our argument that inclusion of damping leads to a complex ket $|g_F^{\mathrm{damp}}\rangle$ even for a nondegenerate ground state, such that $|g_F^{\mathrm{damp}}\rangle$ is linearly independent of $|g_F^{\mathrm{damp}}\rangle^*$, which is incompatible with nondegeneracy.

We conclude that damping factors must not be incorporated in the perturbation of the ground state by a static field.

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