Comment on "Suppression of Angular-Momentum Mixing in Photoexcitation of Rydberg States by Multistate Off-Resonant Quantum Coherence"

In a recent Letter [1], Muller and Noordam discuss the angular-momentum mixing in photoexcitation of Rydberg states in what they refer to as an extension of an essential state model originally introduced by Corless and Stroud [2]. The later model only includes one degenerate (the resonant) Rydberg manifold. The essential state model of Ref. [1] includes also other degenerate states lying outside the bandwidth of the driving laser. Muller and Noordam show that the angular mixing predicted by the model of Ref. [2] can be strongly suppressed by multistate quantum coherence due to the very small fraction of the total population accommodated by off-resonant states.

A careful examination of the model presented in Ref. [1] compels me to point out what seems to be a built-in inconsistency. In solving their model, the authors start out with a diagonalization of the laser-electron interaction, V_L, within the degenerate "spherical" $|nl\rangle$ states of one "principal" quantum number n. The eigenfunctions are the "parabolic" states, $|nk\rangle = \sum_l S_{kl}|nl\rangle$, where the coefficients S_{kl} define a unitary transformation. The crucial point, on which the solution of the model and therefore the conclusions are based, is the statement that (1) "this transformation not only removes the coupling between states of the same n, but actually decouples *all* states of differing k" [see also Eq. (5) of Ref. [1]]. If now, e.g., the ground state is chosen to mimic the situation in hydrogen, i.e., as a nondegenerate state, say, $|g\rangle = |n = 1, l = 0\rangle$ which corresponds to the state $|n = 1, k = 0\rangle$ in the parabolic basis, the statement (1) implies that $|g\rangle$ couples only to parabolic states with $k = 0$. This is in conflict with the fact (which is also used in Ref. [1]) that $|g\rangle$ couples to all spherical states with $l = 1$ and therefore to all parabolic states with a coupling depending on the Rabi frequency and the k-dependent transformation coefficients S_{kl}. The coupling between parabolic states with differing principal quantum numbers n is therefore in general not diagonal in k. In the case of the pure Coulomb potential, Bethe and Salpeter give the exact expression on page 276 in their book [3]. As an example, to illustrate my point further, I draw the attention to the related and well-known linear Stark effect in hydrogen where the parabolic states diagonalize the field interaction within *one* principal shell but certainly do not decouple states of differing k belonging to different principal shells.

The remaining discussion and the analytical derivation in Ref. [1] rest heavily on the assumption of a diagonal coupling in k between parabolic states. Accordingly, the model cannot—*a priori*—be expected to predict the correct behavior of angular-momentum mixing in photoexcitation of Rydberg states. I also note that the predictions of the model are *insensitive* to the wavelength of the driving field which seems contrary to what one would expect on physical grounds. Although this drawback may not be very serious in the context of Ref. [1], it would lead to questionable predictions if applied to more general schemes involving more than one wavelength [4]. Finally, I note that the dipole approximation, on which the discussion of Ref. [2], and as a consequence also that of Ref. [1], is based, is a very poor approximation at the considered wavelengths for the Rydberg states involved [4]. In this sense, the mathematical problem of excitation and angular-momentum mixing under those conditions is still open. The theoretical prediction of the outcome of an experimental implementation would require all the details of the specific excitation scenario.

Since various forms of essential state models are employed in a broader context of problems, I feel it is of interest to the general readership to have the shortcomings of such models pointed out.

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