Higher-order susceptibilities of Rydberg-state diamagnetic manifold: discontonuities between degenerate and equidistant spectra

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The interaction between atoms and magnetic field provides an important information concerning atomic structure and behaviour of matter in the field. Atoms in Rydberg states are of particular interest, because their behaviour in moderate fields, available in laboratories, represents behaviour of the total atomic spectrum in strong and superstrong magnetic fields. The consecutive transition to higher levels in a fixed field enables one to clarify the transformations from regularity to chaos for an atomic level when the field strength gradually increases.

The diamagnetic interaction becomes of importance for Rydberg atoms in rather weak fields when the perturbation theory for the description of the energy level shift and splitting applies. However, the first-order diamagnetic energy corrections provide only preliminary information on the Rydberg spectrum in the field. That is why the calculations of higher-order corrections contribute essentially to understand the intrinsic properties of diamagnetic manifold with a given magnetic quantum number m.

For great majority of atoms the Zeeman manifold with $|m| > 3$ is hydrogenlike since it consists of states with large orbital quantum numbers $l \geq |m|$ that have no quantum defects. Therefore the properties of diamagnetic states with $m > 3$ in hydrogen are of a general interest and may be used to describe the Rydberg spectrum of arbitrary atom in external field.

Unlike the Stark manifold, where higher-order corrections were derived in a closed form as polynomials of parabolic quantum numbers [1], for Zeeman effect the diamagnetic corrections may be derived by straightforward diagonalization but only for the first-order and second-order effective hamiltonians that use corresponding integrals of motion in a subspace with a given principal quantum number n [2].

We have proposed a method based on the use of the reduced Green function within a complete basis of hydrogenlike states (including continuum) that makes it possible to solve the secular systems of equations for higher-order corrections to the wave function and energy of degenerate states [3]. All the relevant diamagnetic matrix elements may be calculated in a closed analytical form up to the third order in diamagnetic interaction with the use of the Sturm-series presentation for the Coulomb Green function. The analytical data was used for numerical calculations of third-order diamagnetic susceptibilities of Zeeman manyfold in hydrogen with $|m| = 0, 1$. The correlation between the higher-order and lower-order susceptibilities of a given diamagnetic state has been confirmed in our calculations. E.g., the degeneracy in parity of the lower-frequency part of a manyfold with fixed n and m (approximately one fifth part of the total energy width that includes one fourth part of the total number of states): the doublet structure that was discovered in the lowest orders of diamagnetic interaction [4] continues in the third-order susceptibilities to a high degree of precision (up to 5-6 decimal digits). The rest of the spectrum are equidistant parity-alternating states in all three orders of diamagnetic susceptibilities available in our calculations.

A new property has been discovered in numerical calculations for the third-order susceptibilities. It consists in abrupt discontinuity in the correlated energy dependence of susceptibilities, that appears at the boundary between the doublet and splitted parts of diamagnetic spectrum. The sharpness of that discontinuity becomes more significant for the higher principal quantum number n (in general, for the higher dimension of space of states) and for the higher order of perturbation theory, so it was not noticeable in the second-order calculations and could be noted only in the third-order ones [3].

In this article we report on the general properties of the correlated dependence between the first-order and higher-order diamagnetic susceptibilities of the Rydberg-state Zeeman manyfold. The discussion will be given for $|m| = 4, 5$-states with extremely high principal quantum number $n \approx 100$.

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