Spectral decomposition at a complex laser polarization configuration*

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We study the role of laser polarization in the diamagnetic spectrum for the transition from the ground state to the highly excited Rydberg states through a single photon absorption. For simplicity, one usually polarizes the irradiation laser to the selected main quantum axis, which is along the applied external electric or magnetic field. The transition selection rule is simply expressed as $\Delta m = 0$, which corresponds to the π transition. When the polarization is circularly polarized around the main axis, the σ^+ or σ^- transition occurs, corresponding to the selection rule of $\Delta m = 1$ or $\Delta m = -1$, respectively. A slightly more complex case is that the laser is linearly polarized perpendicular to the main axis. The numerical calculation shows that we can decompose the transition into the sum of σ^+ and σ^- transitions, it is noted as the σ transition. For the more complex case in which the laser is linearly polarized with an arbitrary angle with respect to the main axis, we have to decompose the polarization into one along the main axis and the other one perpendicular to the main axis. They correspond to π and σ transitions, respectively. We demonstrate that these transitions in the diamagnetic spectrum and the above spectral decomposition well explain the experimentally observed spectra.

Keywords: diamagnetic spectrum, Rydberg atoms, laser polarization

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1. Introduction

Spectrometry is one of the most basic and efficient tools to reveal atomic and molecular structures[1,2] and even to supervise complex molecular dynamics such as the forming of medicine.[3] For atoms in free external fields or molecules without alignment, the optical transition actually happens with a large amount of degenerate states, and thus the geometry of the laser polarization is not so important. However, for those atoms and molecules in an external field, the external field will polarize the atoms or align the molecules to define the so-called main quantum axis, usually denoted as the z axis. In this case, the laser polarization plays an important role in the spectral profile.

The optical transition can be described with the aid of the time-independent Schrödinger equation. The Schrödinger equation is

$$(H+V)|f\rangle = E|f\rangle, \tag{1}$$

where H is the Hamiltonian of an atom or molecule in zero-field, V is the contribution of the interaction of external field with atom or molecule, and $|f\rangle$ is the eigenvector corresponding to eigenvalue E. The oscillator strength for the one-photon dipole transition from initial state $|i\rangle$ to final eigenstate $|f\rangle$ can be expressed as

$$c_{i\to f} = |\langle i|\hat{\boldsymbol{n}}\cdot\boldsymbol{d}|f\rangle|^2, \tag{2}$$

where $\boldsymbol{d}$ is the electric dipole operator, and $\hat{\boldsymbol{n}}$ is the unit vector along the laser electric field. Given an initial state and the dipole moment, if we solve the eigenvector in Eq. (1), the spectral information can be determined accordingly.

Usually the dipole is aligned along the external field, and the z axis is defined along this direction. The laser used to excite the atom or molecule is closely correlated with the dipole vector. If the laser is linearly polarized along the z axis, the transition will not change the magnetic quantum number of the concerned state. If a left- circular or right-circular laser is used, the transition will increase or decrease the magnetic quantum number by one. These transitions are the basic optical processes corresponding to operators d_z, d_+, and d_-. They lead to the simplest spectra corresponding to π, σ^+, and σ^- transitions, respectively. However, if the laser is polarized into other directions, the transition is very complicated, leading to a complex spectral structure.

The spectrum for a complex laser polarization configuration is not preferable, since it is complicated. However, sometimes, one cannot escape this morass due to the constraints of the actual experimental apparatus.[4–7] It is also interesting to study more general polarization arrangements although these cause significant complications for the interpretation of experimental spectra. Moreover, the time saving could be significant since the spectrum obtained in a single laser scan can contain information from all of the basic transitions π, σ^+, and σ^-. In this study, the laser polarization is aligned with the magnetic field at an angle of 30°.

In this paper, we demonstrate these complex polarization effects on the diamagnetic spectrum though the diamagnetism has been experimentally studied,[8,9] especially by the group of Connerade,[10–14] as well as in our laboratory.[15–17] In the calculations, the exact quantum defect theory[18] and the complex

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coordinate rotation[19] incorporating the B-Spline basis[20,21] are used.

The experiment uses the same apparatus described previously,[16,22] except for a more flexible laser polarization configuration. As in the previous work, the atomic beam propagates along the magnetic field direction which eliminates the motional Stark effect, while the laser beam is aligned perpendicular to the magnetic field so the Doppler broadening is minimized. Under this configuration, we can arrange the laser polarization parallel or perpendicular to or even at an arbitrary angle with respect to the magnetic field, thus it is possible for us to study the dependence of the diamagnetic spectrum on the laser polarization.

2. Theory of spectral decomposition

As is well known, there are three basic polarization types π, σ^+, and σ^-, corresponding to the basic transitions $\Delta m = 0$, $\Delta m = 1$, and $\Delta m = -1$, respectively, as shown in Fig. 1(a). For simplifying the spectral analysis, experimentalists usually excite the atom samples under these polarization configurations.

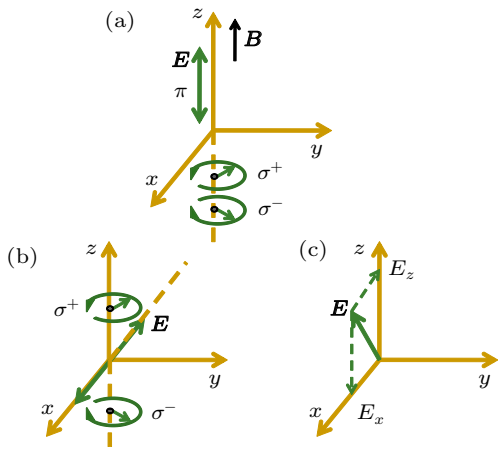


Fig. 1. (color online) Decomposition of the laser polarization. (a) There are three basic polarization types π, σ^+, and σ^-, corresponding to the basic transitions $\Delta m = 0$, $\Delta m = 1$, and $\Delta m = -1$, respectively. (b) The linear polarization perpendicular to the z axis can be decomposed into the sum of equally weighted σ^+ and σ^-, which is denoted as σ by convention. (c) The laser is polarized at an arbitrary angle with respect to the z axis.

Constrained by the experimental apparatus,[4–7] sometimes, the laser can not propagate along the z axis, resulting in the failure of applying the σ^+ or σ^- polarization. One has to linearly polarize the laser parallel or perpendicular to the z axis. For the latter case, the linear polarization can be decomposed into the sum of equally weighted σ^+ and σ^-, which is denoted as σ by convention, as shown in Fig. 1(b).

The more complex case is that the laser is polarized at an arbitrary angle with respect to the z axis, as shown in Fig. 1(c). In this case, we can project it into one component of π along the z axis and one compound component of σ perpendicular to the z axis. Although this configuration will complicate the spectral analysis, it can provide the full physical information. We will demonstrate this spectral decomposition in the next part by combining it with the experimental data.

3. Results and discussion

To show the above spectral decomposition method, in the experiments, following the polarization configurations in Fig. 1, we polarize the laser linearly along or perpendicular to the magnetic field direction, and even at an arbitrary angle with respect to the magnetic field, and observe the corresponding spectra. The single photon absorption spectrum covers an energy range extending from -140 cm^{-1} to -70 cm^{-1} below the ionization threshold, and the magnetic field applied varies from 1.5 T to 3 T. For a better visual comparison with the experimental data, the calculated spectra are convoluted with a Gaussian function of width 0.15 cm^{-1} and displayed upside-down in mirror fashion.

3.1. Laser linearly polarized along the magnetic field

The $\Delta m = 0$ diamagnetic spectrum is obtained when the laser photon is linearly polarized along the magnetic field. The spectra under various magnetic fields are shown in Fig. 2. Under low magnetic fields, i.e., at $B = 2$ T, the l-mixing can not be resolved clearly at low energy, while it is possible in the higher energy region, for instance, at $E = -75$ cm^{-1}, as shown in Fig. 2(a). With the increase of the magnetic field intensity, as shown in Figs. 2(b) and 2(c), different l states with the same parity more strongly interact with each other due to the diamagnetic effect, the spectral lines become fully resolved. At $B = 3$ T, as shown in Fig. 2(c), different n-manifolds intrude into each other, and the spectrum displays n-mixing. The calculation is similar to the σ^+ spectrum obtained previously,[16]

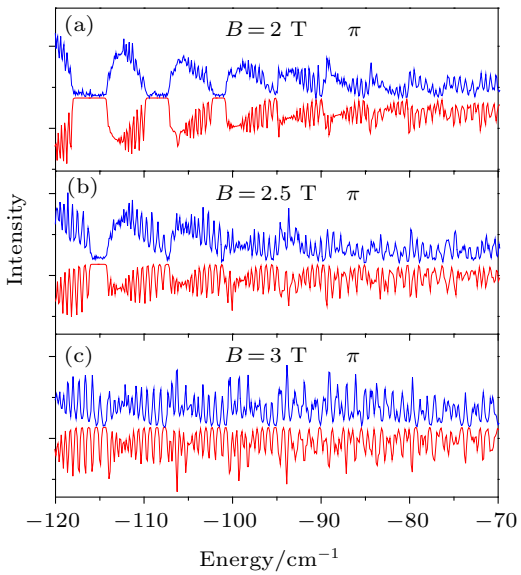


Fig. 2. (color online) Observed (upward ticking in blue) and calculated (downward ticking in red) π diamagnetic spectra of barium. The π corresponds to the transition from the $m = 0$ ground state to the $m = 0$ excited states without changing the magnetic quantum number m, which is determined by the laser polarization along the magnetic field. It is one of the simplest transition types widely employed in experiments. Various magnetic fields ((a) $B = 2$ T, (b) $B = 2.5$ T, (c) $B = 3$ T) are used.

but the laser is linearly polarized along the magnetic field, permitting only $\Delta m = 0$ transition to occur. The excited Rydberg states have the same magnetic quantum number with the ground state, i.e., $m = 0$, since the ground state is $6s^2\,{}^1S_0$. As one of the three basic polarization types, the π polarized diamagnetic spectra in Fig. 2 show more complex structures than the σ^+ and σ^- ones.[16]

3.2. Laser linearly polarized perpendicular to the magnetic field

When the excitation laser is propagating along the magnetic field and is circular polarized, the $\Delta m = 1$ or $\Delta m = -1$ transitions take place, corresponding respectively to σ^+ and σ^- spectra. This case has been well studied before.[13,16,23–25] What happens if the laser field is linearly polarized perpendicular to the magnetic field? One is obliged to take this kind of excitation scheme only when limited by the geometrical constraints of the apparatus.[4–6] Nevertheless, from a theoretical point of view this could be interesting, as the group of states with $m = 1$ and $m = -1$ do not interact at all because m is a good quantum number.[4] Thus both σ^+ and σ^- transitions will occur, resulting in two identical sets of spectra in one spectral observation, although they can be offset by the energy corresponding to the paramagnetic interaction with magnitude of B in atomic units. It is denoted by σ conventionally.[4–6]

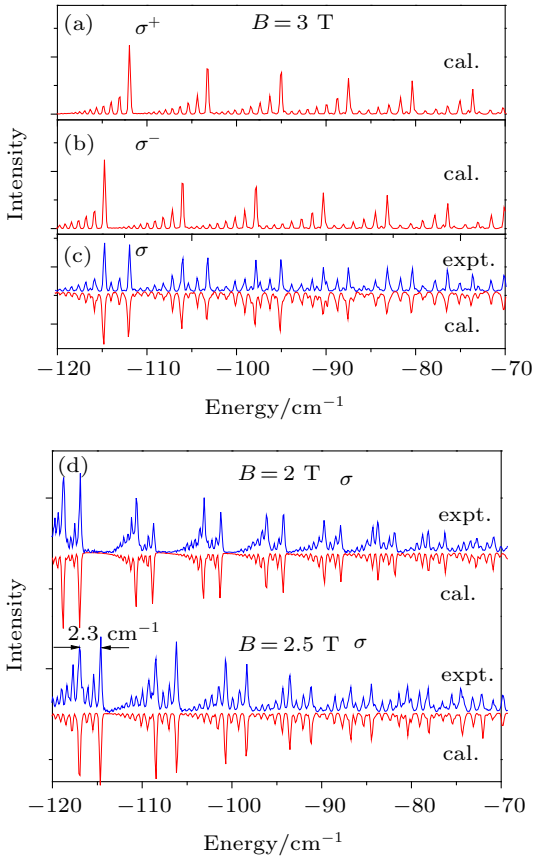


Fig. 3. (color online) Buildup of σ polarized diamagnetic spectrum. It is demonstrated for the spectrum under a magnetic field of $B = 3$ T. The results for (a) σ^+ and (b) σ^- components are calculated separately, and their equally weighted sum is shown (downward ticking in red) in the lower part of panel (c), which agrees well with the experimental measurement (upward ticking in blue). (d) Calculated compound spectra for other magnetic fields, all of them are in coincidence with the experiments well.

The above discussion suggests a simple approach for the theoretical interpretation of the spectra. Basically one can decompose the linear polarization perpendicular to the magnetic field into two equally-weighted circular polarizations σ^+ and σ^-, and the final σ spectrum is therefore a weighted sum of the σ^+ and σ^- transition spectra. The formation of the σ diamagnetic spectrum under a magnetic field of $B = 3$ T is demonstrated in Fig. 3(a)–3(c). More comparisons between the calculated and the observed results are shown in Fig. 3(d). In all the discussed cases, we find good agreement between theory and experiment. As an example, we confirm that the energy offset between σ^+ and σ^- is about 2.3 cm^{-1}, as shown Fig. 3(d), corresponding to the paramagnetic energy difference at $B = 2.5$ T. Since the σ spectra are composed of essentially identical σ^+ and σ^- ones except for the offset, the detailed diamagnetic structures for the σ polarization can be deduced from the σ^+ polarized ones.[16]

3.3. Laser linearly polarized at an arbitrary angle to the magnetic field

We next turn to the more complicated case of a linearly polarized laser in an arbitrary direction with respect to the magnetic field. To our knowledge, this has not been experimentally studied previously and no theoretical calculation exists.

We assume that the magnetic field is along the z axis and the laser polarization makes an angle θ with the z axis. A natural approach to study the spectrum is decomposing the electric vector $\boldsymbol{E}$ of the laser field into a parallel component $\boldsymbol{E}_z$ along the magnetic field and a perpendicular component $\boldsymbol{E}_x$ along the x axis. They satisfy the relations

$$\boldsymbol{E}_z = |E|\cos\theta\,\hat{\boldsymbol{z}}, \tag{3a}$$

$$\boldsymbol{E}_x = |E|\sin\theta\,\hat{\boldsymbol{x}}. \tag{3b}$$

The component perpendicular to the magnetic field can be further decomposed into a superposition of the left and the right circular polarizations as previously discussed. In the perpendicular plane, the electric vector $\boldsymbol{E}_x$ can be expressed as

$$\begin{aligned}\boldsymbol{E}_x &= E_x\cos\omega t\,\hat{\boldsymbol{x}}\\ &= \frac{1}{\sqrt{2}}E_x\Big[\frac{1}{\sqrt{2}}\Big(\cos\omega t\,\hat{\boldsymbol{x}} + \cos\Big(\omega t + \frac{\pi}{2}\Big)\hat{\boldsymbol{y}}\Big)\\ &\quad + \frac{1}{\sqrt{2}}\Big(\cos\omega t\,\hat{\boldsymbol{x}} + \cos\Big(\omega t - \frac{\pi}{2}\Big)\hat{\boldsymbol{y}}\Big)\Big].\end{aligned} \tag{4}$$

In other words, this linear polarization is decomposed into two equally-weighted σ^+ and σ^-. The intensity ratio R of σ to π is

$$R = \tan^2\theta/2. \tag{5}$$

In our experiment, θ is set to 30° and the diamagnetic spectrum with laser polarization in an arbitrary direction is demonstrated. As shown in Fig. 4, the spectra of π and σ transitions are calculated separately and shown in Figs. 4(a) and

4(b), and they are averaged into the upper spectrum in Fig. 4(c) accordingly to the intensity ratio $R_{\sigma/\pi} = \tan^2\theta/2 \sim 0.17$. The weighted average spectrum agrees well with the experimental spectrum shown in the lower part of Fig. 4(c).

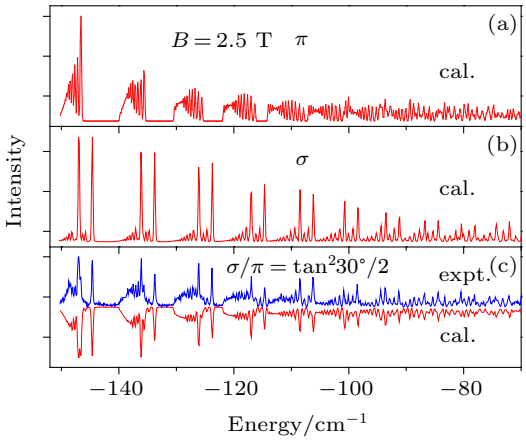


Fig. 4. (color online) Spectral decomposition of the diamagnetic spectrum with laser polarization at an angle $\theta = 30^\circ$ with respect to the magnetic field. The spectra for (a) π and (b) σ are calculated separately, and their weighted composition according to the intensity ratio $R_{\sigma/\pi} = \tan^2\theta/2 \sim 0.17$ is shown (downward ticking in red) in panel (c). It agrees well with the experiment.

4. Conclusion

The diamagnetic spectra of barium atoms are studied experimentally at several excitation configurations. They are compared with theoretical calculations including atomic core effects, and the agreements are satisfying. In the theoretical calculations, for the laser polarization perpendicular to the magnetic field, the spectrum is decomposed into left- and right-circular polarized components, while for an arbitrary polarization, the spectrum is obtained from a weighted sum of a linear polarization component parallel to the magnetic field and a component perpendicular to the magnetic field. When two excited states with different m are adjacent in energy, we can choose an appropriate polarization direction to populate them simultaneously with a specified ratio. A near-resonant microwave can further couple these two states, allowing for internal state manipulations. Our calculation provides a universal method capable of reproducing the diamagnetic spectrum with the laser polarization aligned to the magnetic field at any angle and within an energy range of interest. In fact, as the diamagnetic spectrum is sensitive to the laser polarization direction, we can use the atomic transition spectrum to determine the polarization direction of a laser.

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