Magnetoexcitons Break Antiunitary Symmetries

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We show analytically and numerically that the application of an external magnetic field to highly excited Rydberg excitons breaks all antiunitary symmetries in the system. Only by considering the complete valence band structure of a direct-band-gap cubic semiconductor, the Hamiltonian of excitons leads to the statistics of a Gaussian unitary ensemble without the need for interactions with other quasiparticles like phonons. Hence, we give theoretical evidence for a spatially homogeneous system breaking all antiunitary symmetries.

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For more than 100 years, one distinguishes in classical mechanics between two fundamentally different types of motion: regular and chaotic motion. Their appearance strongly depends on the presence of underlying symmetries, which are connected with constants of motion and reduce the degrees of freedom in a given system. If symmetries are broken, the classical dynamics often become nonintegrable and chaotic. However, since the description of chaos by trajectories and Lyapunov exponents is not possible in quantum mechanics, it has been unknown for a long time how classical chaos manifests itself in quantum mechanical spectra [1,2].

The Bohigas-Giannoni-Schmit conjecture [3] suggests that quantum systems with few degrees of freedom and with a chaotic classical limit can be described by random matrix theory [4,5] and thus, show typical level spacings. At the transition to quantum chaos, the level spacing statistics will change from Poissonian statistics to the statistics of a Gaussian orthogonal ensemble (GOE) or a Gaussian unitary ensemble (GUE) as symmetry reduction leads to a correlation of levels and hence, to a strong suppression of crossings [1].

To which of the two universality classes a given system belongs is determined by remaining antiunitary symmetries in the system. While GOE statistics can be observed in many different systems like, e.g., in atomic [6,7] and molecular spectra [8], for nuclei in external magnetic fields [9–12], microwaves [13–15], impurities [16], and quantum wells [17], GUE statistics appears only if *all* antiunitary symmetries are broken [3,18]. Thus, GUE statistics are observable only in very exotic systems like microwave cavities with ferrite strips [19] or billards in microwave resonators [20] and graphene quantum dots [21].

There is no example for a system showing GUE statistics in atomic physics. This is especially true for one of the prime examples when studying quantum chaos: the highly excited hydrogen atom in strong external fields. Even though the applied magnetic field breaks time-reversal invariance, at least one antiunitary symmetry, e.g., time reversal and a certain parity, remains and GOE statistics are observed [1,22,23].

Excitons are fundamental quasiparticles in semiconductors, which consist of an electron in the conduction band and a positively charged hole in the valence band. Recently, T. Kazimierczuk *et al.* [24] have shown in a remarkable high-resolution absorption experiment an almost perfect hydrogenlike absorption series for the yellow exciton in cuprous oxide (Cu_2O) up to a principal quantum number of $n = 25$. This experiment has drawn new interest to the field of excitons experimentally and theoretically [25–35].

Since excitons in semiconductors are often treated as the hydrogen analog of the solid state, but also show substantial deviations from this behavior due to the surrounding solid, the question about their level spacing statistics in external fields arises. First experimental investigations of the level spacing statistics in an external magnetic field give indications on a breaking of antiunitary symmetries, which is, however, attributed to the interaction of excitons and phonons [31].

Very recently, we have shown that it is indispensable to account for the complete valence band structure of Cu_2O in a quantitative theory of excitons [28] to explain the striking experimental findings of a fine structure splitting and the observability of F excitons [25]. We have also proven that the effect of the valence band structure on the exciton spectra is even more prominent when treating excitons in external fields [35].

In this Letter, we will now show that the simultaneous presence of a cubic band structure and external fields will break all antiunitary symmetries in the exciton system without the need of phonons. This effect is present in all direct-band-gap semiconductors with a cubic valence band structure and not restricted to Cu_2O. We prove not only analytically that the antiunitary symmetry known from the hydrogen atom in external fields is broken in the case of excitons, but also, by solving the Schrödinger equation in a complete basis, that the nearest-neighbor spacing distribution of exciton states reveals GUE statistics. Thus, we give the first theoretical evidence for a spatially homogeneous system which breaks all antiunitary symmetries, and demonstrate a fundamental difference between atoms in vacuum and excitons.

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Without external fields, the Hamiltonian of excitons in direct-band-gap semiconductors reads [28]

$$H = E_g - e^2/4\pi\varepsilon_0\varepsilon|\boldsymbol{r}_e - \boldsymbol{r}_h| + H_e(\boldsymbol{p}_e) + H_h(\boldsymbol{p}_h), \quad (1)$$

with the band gap energy E_g, the Coulomb interaction, which is screened by the dielectric constant ε, and the kinetic energies of electron and hole. While the conduction band is almost parabolic in many semiconductors, and thus, the kinetic energy of the electron can be described by the simple expression $H_e(\boldsymbol{p}_e) = \boldsymbol{p}_e^2/2m_e$, with the effective mass m_e, the kinetic energy of the hole in the case of three coupled valence bands is given by the more complex expression [26,28,36]

$$\begin{aligned} H_h(\boldsymbol{p}_h) = H_{\text{so}} &+ (\gamma_1 + 4\gamma_2)\boldsymbol{p}_h^2/2m_0 \\ &- 3\gamma_2(p_{h1}^2 \boldsymbol{I}_1^2 + \text{c.p.})/\hbar^2 m_0 \\ &- 6\gamma_3(\{p_{h1}, p_{h2}\}\{\boldsymbol{I}_1, \boldsymbol{I}_2\} + \text{c.p.})/\hbar^2 m_0, \quad (2) \end{aligned}$$

with $\{a, b\} = (ab + ba)/2$. Here, γ_i denotes the three Luttinger parameters, m_0 the free electron mass, and c.p. cyclic permutation. The threefold degenerate valence band is accounted for by the quasispin $I = 1$, which is a convenient abstraction to denote the three orbital Bloch functions xy, yz, and zx [36]. The components of its matrices $\boldsymbol{I}_i$ are given by $I_{i,jk} = -i\hbar\varepsilon_{ijk}$ [28,36], with the Levi-Civita symbol ε_{ijk}. Note that the expression for $H_h(\boldsymbol{p}_h)$ can be separated in two parts having spherical and cubic symmetry, respectively [37]. The coefficients μ' and δ' of these parts can be expressed in terms of the three Luttinger parameters: $\mu' = (6\gamma_3 + 4\gamma_2)/5\gamma_1'$ and $\delta' = (\gamma_3 - \gamma_2)/\gamma_1'$, with $\gamma_1' = \gamma_1 + m_0/m_e$ [28,37,38]. Finally, the spin-orbit coupling $H_{\text{so}} = 2\Delta/3(1 + \boldsymbol{I}\cdot\boldsymbol{S}_h/\hbar^2)$ between I and the hole spin S_h describes a splitting of the valence bands at the center of the Brillouin zone [38].

Let us now consider the case with external fields being present. Then, the corresponding Hamiltonian is obtained via the minimal substitution. We further introduce relative and center of mass coordinates and set the position and momentum of the center of mass to zero [39,40]. Then, the complete Hamiltonian of the relative motion reads [41–45]

$$\begin{aligned} H = E_g &- e^2/4\pi\varepsilon_0\varepsilon|\boldsymbol{r}| + H_B + e\Phi(\boldsymbol{r}) \\ &+ H_e[\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r})] + H_h[-\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r})], \quad (3) \end{aligned}$$

with the relative coordinate $\boldsymbol{r} = \boldsymbol{r}_e - \boldsymbol{r}_h$ and the relative momentum $\boldsymbol{p} = (\boldsymbol{p}_e - \boldsymbol{p}_h)/2$ of electron and hole [39,40].

Here, we use the vector potential $\boldsymbol{A} = (\boldsymbol{B}\times\boldsymbol{r})/2$ of a constant magnetic field B and the electrostatic potential $\Phi(\boldsymbol{r}) = -\boldsymbol{F}\cdot\boldsymbol{r}$ of a constant electric field F. The term H_B describes the energy of the spins in the magnetic field [36,42,45,46]. In this Letter, we want to show that the Hamiltonian (3) breaks all antiunitary symmetries. Since the term H_B, as well as the spin-orbit interaction, are invariant under the symmetry operations considered below, we will neglect them in the following.

Before we investigate the symmetry of H, we have to note that the matrices $\boldsymbol{I}_i$ are not the standard spin matrices $\boldsymbol{S}_i$ of spin one [47]. However, since these matrices obey the commutation rules [36]

$$[\boldsymbol{I}_i, \boldsymbol{I}_j] = i\hbar\sum_{k=1}^{3}\varepsilon_{ijk}\boldsymbol{I}_k, \quad (4)$$

there must be a unitary transformation $\boldsymbol{U}$ such that $\boldsymbol{U}^\dagger\boldsymbol{I}_i\boldsymbol{U} = \boldsymbol{S}_i$ holds. This transformation matrix reads

$$\boldsymbol{U} = \frac{1}{\sqrt{2}}\begin{pmatrix} -1 & 0 & 1 \\ -i & 0 & -i \\ 0 & \sqrt{2} & 0 \end{pmatrix}, \quad (5)$$

and we will now use the matrices $\boldsymbol{S}_i$ instead of $\boldsymbol{I}_i$ in the following.

In the special case with vanishing Luttinger parameters $\gamma_2 = \gamma_3 = 0$, the exciton Hamiltonian (3) is of the same form as the Hamiltonian of a hydrogen atom in external fields. It is well known that for this Hamiltonian, there is still one antiunitary symmetry left, i.e., that it is invariant under the combined symmetry of time inversion K, followed by a reflection $S_{\hat{\boldsymbol{n}}}$ at the specific plane spanned by both fields [1]. This plane is determined by the normal vector

$$\hat{\boldsymbol{n}} = (\boldsymbol{B}\times\boldsymbol{F})/|\boldsymbol{B}\times\boldsymbol{F}| \quad (6)$$

or $\hat{\boldsymbol{n}}\perp\boldsymbol{B}/B$ if $F = 0$ holds. Therefore, the hydrogenlike system shows GOE statistics in the chaotic regime.

As the hydrogen atom is spherically symmetric in the field-free case, it makes no difference whether the magnetic field is oriented in z direction or not. However, in a semiconductor with $\delta' \neq 0$, the Hamiltonian has cubic symmetry and the orientation of the external fields with respect to the crystal axis of the lattice becomes important. Any rotation of the coordinate system with the aim of making the z axis coincide with the direction of the magnetic field will also rotate the cubic crystal lattice. Hence, we will show that the only remaining antiunitary symmetry mentioned above is broken for the exciton Hamiltonian if the plane spanned by both fields is *not* identical to one of the symmetry planes of the cubic lattice. Even without an external electric field, the symmetry is broken if the magnetic field is not oriented in one of these symmetry planes. Only if the plane spanned by both fields is identical to one of the symmetry planes of the cubic lattice, the antiunitary symmetry $KS_{\hat{\boldsymbol{n}}}$, with $\hat{\boldsymbol{n}}$ given by Eq. (6), is present.

At first, we will show this analytically. Under time inversion K and reflections $S_{\hat{\boldsymbol{n}}}$, at a plane perpendicular to a normal vector $\hat{\boldsymbol{n}}$, the vectors of position $\boldsymbol{r}$, momentum $\boldsymbol{p}$, and spin $\boldsymbol{S}$ transform according to [47]

$$KrK^\dagger = \boldsymbol{r}, \qquad K\boldsymbol{p}K^\dagger = -\boldsymbol{p}, \qquad K\boldsymbol{S}K^\dagger = -\boldsymbol{S}, \tag{7}$$

and

$$S_{\hat{n}}\boldsymbol{r}S_{\hat{n}}^\dagger = \boldsymbol{r} - 2\hat{\boldsymbol{n}}(\hat{\boldsymbol{n}}\cdot\boldsymbol{r}), \tag{8a}$$

$$S_{\hat{n}}\boldsymbol{p}S_{\hat{n}}^\dagger = \boldsymbol{p} - 2\hat{\boldsymbol{n}}(\hat{\boldsymbol{n}}\cdot\boldsymbol{p}), \tag{8b}$$

$$S_{\hat{n}}\boldsymbol{S}S_{\hat{n}}^\dagger = -\boldsymbol{S} + 2\hat{\boldsymbol{n}}(\hat{\boldsymbol{n}}\cdot\boldsymbol{S}). \tag{8c}$$

Let us denote the orientation of $\boldsymbol{B}$ and $\boldsymbol{F}$ in spherical coordinates via $\boldsymbol{B}(\varphi,\vartheta) = B(\cos\varphi\sin\vartheta, \sin\varphi\sin\vartheta, \cos\vartheta)^T$.

Possible orientations of the fields breaking the antiunitary symmetry are then, e.g., $\boldsymbol{B}(0,0)$ and $\boldsymbol{F}(\pi/6,\pi/2)$, $\boldsymbol{B}(0,\pi/6)$ and $\boldsymbol{F}(\pi/2,\pi/2)$, or $\boldsymbol{B}(\pi/6,\pi/6)$ and $\boldsymbol{F}=\boldsymbol{0}$. In all of these cases, the hydrogenlike part of the Hamiltonian (3) is invariant under $KS_{\hat{n}}$, with $\hat{\boldsymbol{n}}$ given by Eq. (6). However, other parts of the Hamiltonian like $H_c = (p_1^2\boldsymbol{S}_1^2 + \text{c.p.})$ [see Eq. (2)] are not invariant. For example, for the case with $\boldsymbol{B}(0,0)$ and $\boldsymbol{F}(\pi/6,\pi/2)$, we obtain

$$\begin{aligned} &S_{\hat{n}}KH_cK^\dagger S_{\hat{n}}^\dagger - H_c \\ &\quad = 1/8[2\sqrt{3}(\boldsymbol{S}_2^2 - \boldsymbol{S}_1^2)p_1p_2 \\ &\qquad + 3(\boldsymbol{S}_1^2p_2^2 + \boldsymbol{S}_2^2p_1^2) - 3(\boldsymbol{S}_1^2p_1^2 + \boldsymbol{S}_2^2p_2^2) \\ &\qquad + \{\boldsymbol{S}_1,\boldsymbol{S}_2\}(2\sqrt{3}(p_2^2 - p_1^2) + 12p_1p_2)] \neq 0, \end{aligned} \tag{9}$$

with $\hat{\boldsymbol{n}} = (-1/2, \sqrt{3}/2, 0)^T$. Thus, the generalized time-reversal symmetry of the hydrogen atom is broken for excitons due to the cubic symmetry of the semiconductor.

Since a breaking of all antiunitary symmetries is connected with the appearance of GUE statistics, we now solve the Schrödinger equation corresponding to H for the arbitrarily chosen set of material parameters $E_g = 0$, $\varepsilon = 7.5$, $m_e = m_0$, $\gamma_1' = 2$, and $\mu' = 0$, using a complete basis. We can then analyze the nearest-neighbor spacings of the energy levels [23]. To reduce the size of our basis and thus, the numerical effort, we already assumed $\Delta = 0$ so that we can disregard the spins of electron and hole.

The cubic part of the Hamiltonian (3) couples the angular momentum L of the exciton and the quasispin I to the total momentum $G = L + I$, with the z component M_G. For the radial part of the basis functions, we use the Coulomb-Sturmian functions of Refs. [28,48], with the radial quantum number N to obtain a complete basis. Hence, the ansatz for the exciton wave function reads

$$|\Psi\rangle = \sum_{NLGM_G} c_{NLGM_G}|N,L,I,G,M_G\rangle, \tag{10}$$

with complex coefficients c.

Without an external electric field, parity is a good quantum number and the operators in the Schrödinger equation couple only basis states with even *or* with odd values of L. Hence, we consider the case with $\boldsymbol{B}(\pi/6,\pi/6)$ and $\boldsymbol{F}=\boldsymbol{0}$ and use only basis states with odd values of L as these exciton states can be observed in direct-band-gap parity-forbidden semiconductors [25,28,29].

After rotating the coordinate system by the Euler angles $(\alpha,\beta,\gamma) = (0,\vartheta,\varphi)$ to make the quantization axis coincide with the direction of the magnetic field [45,49], we write the Hamiltonian in terms of irreducible tensors [37,49]. Inserting the ansatz (10) in the Schrödinger equation $H\Psi = E\Psi$ and multiplying from the left with the state $\langle N', L', I, G', M_G'|$, we obtain a matrix representation of the Schrödinger equation of the form $\boldsymbol{Dc} = E\boldsymbol{Mc}$. The vector $\boldsymbol{c}$ contains the coefficients of the ansatz (10) and the matrix elements entering the matrices $\boldsymbol{D}$ and $\boldsymbol{M}$ can be calculated using the relations given in Ref. [28]. The generalized eigenvalue problem is finally solved using an appropriate LAPACK routine [50]. In our numerical calculations, the maximum number of basis states used is limited by the condition $N + L \leq 29$ due to the required computer memory.

Before analyzing the nearest-neighbor spacings, we have to unfold the spectra to obtain a constant mean spacing [1,3,23,51]. The number of level spacings analyzed is comparatively small since the magnetic field breaks all symmetries in the system and limits the convergence of the solutions of the generalized eigenvalue problem with high energies [28]. As in Ref. [23], we furthermore have to leave out a certain number of low-lying sparse levels to remove individual but nontypical fluctuations. Hence, we use about 250 exciton states for our analysis. Owing to this number of states, we do not present histograms of the level-spacing probability distribution function $P(s)$, but calculate the cumulative distribution function [52]

$$F(s) = \int_0^s P(x)dx. \tag{11}$$

The function $F(s)$ is shown in Fig. 1 for increasing values of the parameter δ' at $B = 3$ T. In this figure, we also show the cumulative distribution function, corresponding to the spacing distributions known from random matrix theory [3,31]: the Poissonian distribution

$$P_P(s) = e^{-s} \tag{12}$$

for noninteracting energy levels, the Wigner distribution

$$P_{\text{GOE}}(s) = \frac{\pi}{2}se^{-\pi s^2/4}, \tag{13}$$

and the distribution

$$P_{\text{GUE}}(s) = \frac{32}{\pi^2}s^2e^{-4s^2/\pi} \tag{14}$$

for systems without any antiunitary symmetry. Note that the most characteristic feature of GUE statistics is the

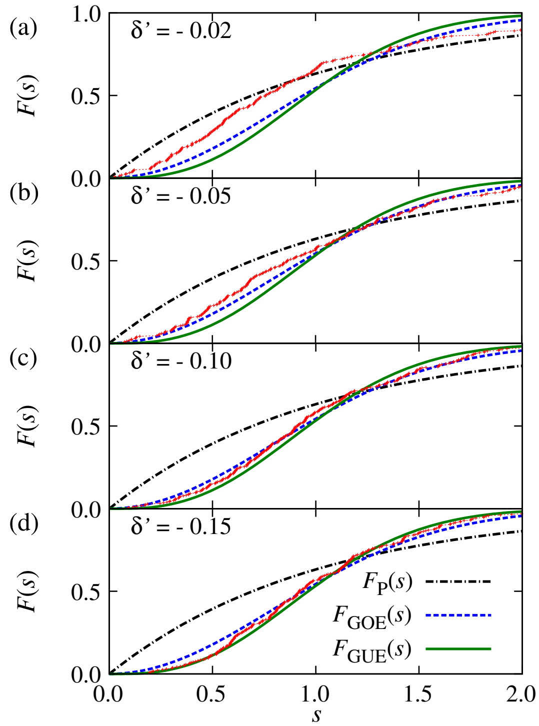

FIG. 1. Cumulative distribution function $F(s)$ for increasing values of δ', with $\boldsymbol{B}(\varphi,\vartheta)=\boldsymbol{B}(\pi/6,\pi/6)$ and $B=3$ T. Besides the numerical data (red dots), we also show the corresponding functions of a Poissonian ensemble (black dash-dotted line), GOE (blue dashed line), and GUE (green solid line). For increasing values of δ', the statistics rapidly change to the one of a Gaussian unitary ensemble (d). Note that we do not show the hydrogenlike case $\delta'=0$ since we simply obtain the transitional form between Poissonian and GOE statistics and since this system is sufficiently well known from literature (see, e.g., Refs. [1,22,23] and further references therein).

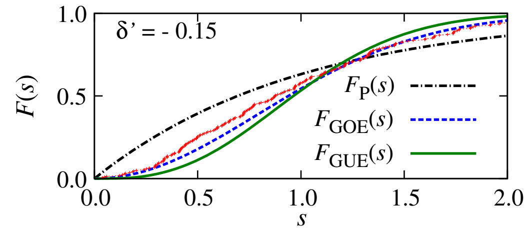

FIG. 2. Cumulative distribution function $F(s)$ for $\delta'=-0.15$, with $\boldsymbol{B}(\varphi,\vartheta)=\boldsymbol{B}(0,\pi/6)$ and $B=3$ T. Since $\boldsymbol{B}$ is oriented in one of the symmetry planes of the lattice, only GOE statistics can be observed when neglecting phonons.

quadratic level repulsion for small s and that the clearest distinction between GOE and GUE statistics can be taken for $0\leq s\lesssim 0.5$. Hence, we see from that, there is clear evidence for GUE statistics. Note that for all results presented in Fig. 1, we used the constant value of $B=3$ T and exciton states within a certain energy range. It is well known from atomic physics that chaotic effects become more apparent in higher magnetic fields or by using states of higher energies for the analysis. Hence, by increasing B or investigating the statistics of exciton states with higher energies, GUE statistics could probably be observed for smaller values of $|\delta'|$. At this point, we have to note that an evaluation of numerical spectra for $\delta'>0$ shows the same appearance of GUE statistics. This is expected since the analytically shown breaking of all antiunitary symmetries is independent of the sign of the material parameters.

If the magnetic field is oriented in one of the symmetry planes of the cubic lattice, only GOE statistics are observable. Indeed, when investigating the exciton spectrum for, e.g., $\boldsymbol{B}(0,\pi/6)$, the level spacing statistics are best described by GOE statistics, especially for small values of s, as can be seen from Fig. 2. Very recently, M. Aßmann *et al.* [31] have shown experimentally that excitons in Cu_2O show GUE statistics in an external magnetic field. However, since their experimental spectra were analyzed exactly for $\boldsymbol{B}(0,\pi/6)$, there must be another explanation for this observation than the cubic band structure. M. Aßmann *et al.* [31] have assigned the observation of GUE statistics to the interaction of excitons and phonons.

The main advantage of theory over the experiments is the fact that the exciton-phonon interaction can be left out. Hence, one can treat the effects of the band structure and of the exciton-phonon interaction separately. We performed model calculations to demonstrate that, in general, GUE statistics appear for a much simpler system; i.e., only the presence of the cubic lattice and the external fields already breaks all antiunitary symmetries without the need for interactions with other quasiparticles like phonons. We did not intend a line-by-line comparison with experimental results. Because of the high dimension of the problem as a result of the presence of the complex band structure, the spin-orbit interaction, and phonons, this is not possible at the moment. However, we do not expect that the effects of the band structure and the phonons on the level spacing statistics will cancel each other out. Indeed, based on the analytic part of our analysis and the fact that the operator describing the interaction between excitons and phonons looks quite different from the operators in our Hamiltonian [44], the phonons certainly do not restore antiunitary symmetries. Instead, the results of Ref. [31] suggest that phonons will further increase the chaos.

We think that Cu_2O is the most promising candidate to investigate the effect of the band structure. As the experiments in Ref. [31] were performed with the magnetic field being oriented in a direction of high symmetry, it would now be highly desirable to investigate exciton absorption spectra in Cu_2O for other orientations of the magnetic field to observe the effect of the band structure on the line statistics.

In conclusion, we have shown analytically and numerically that the cubic symmetry of the lattice and the band structure leads to a breaking of all antiunitary symmetries in the system of magnetoexcitons. This effect demonstrates a fundamental difference between atoms in vacuum and excitons and is not limited to certain values of the material parameters, for which reason it appears in all direct-band-gap semiconductors with a cubic valence band structure. Furthermore, a closer investigation of excitons in external fields can lead to a better understanding of the connection between quantum and classical chaos.

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