Renormalization of the 3D exciton spectrum by the disorder

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Effect of short-range disorder on the excited states of the exciton is studied. Disorder causes an obvious effect of broadening. Microscopically, an exciton, as an entity, is scattered by the large-scale disorder fluctuations. Much less trivial is that short-scale fluctuations, with a period of the order of the Bohr radius, cause a well-defined down-shift of the exciton levels. We demonstrate that this shift exceeds the broadening parametrically and study the dependence of this shift on the orbital number. Difference of the shifts for neighboring levels leads to effective renormalization of the Bohr energy. Most remarkable effect is the disorder-induced splitting of S and P exciton levels. The splitting originates from the fact that disorder lifts the accidental degeneracy of the hydrogen-like levels. The draw an analogy between this splitting and the Lamb shift in quantum electrodynamics.

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I. INTRODUCTION

Early measurements\cite{1,2} of optical absorption in Cu$_2$O crystal films have revealed a series of lines below the main absorption edge. Remarkably, the positions of these lines could be fitted with a hydrogen-like spectrum with high accuracy. For this reason, the series were identified with ground and excited states of the exciton.

Interest to highly excited (Rydberg) states of excitons has been revived after the publication of the paper Ref.\cite{4}. In this paper, the existence of Rydberg excitons in Cu$_2$O with principle quantum number up to $n = 25$ was established on the basis of the high-resolution absorption measurements. Observability of the Rydberg states in Cu$_2$O is explained by the high value of the Bohr energy in this crystals, namely, 94meV\cite{4}.

In subsequent studies, see e.g. Refs.\cite{5,6,7} many peculiarities of Rydberg excitons were revealed via the evolution of the absorption lines with electric field. In particular, the deviation of the binding energies from $\frac{1}{n}$ dependence was related to non-parabolicity of the underlying bandstructure. Concerning the shapes of the absorption lines, it was found to be Lorentzian\cite{4} for $n < 10$ and evolves towards Gaussian with increasing $n$. The widths of the lines decreased with $n$ as $n^{-3}$ and saturated for $n > 10$. It was concluded that the origin of the low-$n$ broadening is radiative recombination, while the higher-$n$ broadening is due to crystal inhomogeneities.

The effect of disorder (inhomogeneities) on Rydberg excitons is an issue of conceptual importance. Indeed, the excited levels of hydrogen possess an accidental degeneracy: $n^2$ wave functions correspond to the level, $n$. This degeneracy is lifted in any given disorder configuration. Thus, inhomogeneous broadening, originating from variation of configurations, is accompanied by splitting, which is also a disorder-induced effect. A nontrivial question arises: which of the two effects is dominant? This question is addressed in the present paper. We consider the simplest model of an exciton in parabolic bands and in the presence of a weak white-noise disorder.

II. DISORDER-INDUCED BROADENING

For simplicity we assume that the disorder potential acts only on the electron. Then the Hamiltonian of the exciton reads

\begin{equation}
H = H_0 + V(r)
\end{equation}
Introducing the center of mass coordinate, $R$, Hamiltonian Eq. (1) in the form

$$\hat{H} = \frac{\hat{p}_e^2}{2m_e} + \frac{\hat{p}_h^2}{2m_h} - \frac{e^2}{\kappa |r_e - r_h|} + V(r_e),$$

(1)

where $m_e$ and $m_h$ are the masses of electron and hole, $\hat{p}_e$ and $\hat{p}_h$ are the corresponding momenta, and $\kappa$ is the dielectric constant. The magnitude of the short-range disorder potential is specified by the correlation relation

$$\langle V(r)V(r') \rangle = \gamma \delta (r - r').$$

(2)

Introducing the center of mass coordinate, $R$, and the relative coordinate, $r$, in a standard way, we rewrite the Hamiltonian Eq. (1) in the form

$$\hat{H} = -\frac{\hbar^2}{2M} \Delta R - \frac{\hbar^2}{2\mu} \Delta r - \frac{e^2}{\kappa r} + V\left(R + \frac{m_h}{M} r\right),$$

(3)

where $M = m_e + m_h$ is the net mass, while $\mu = \frac{m_e m_h}{M}$ is the reduced mass of the exciton. To find the width of the exciton line qualitatively, we first assume that characteristic $R = R_L$ is much bigger than the Bohr radius

$$a = \frac{\hbar^2 \kappa}{\mu e^2}.$$  

(4)

We will later inspect this assumption.

Characteristic magnitude $V = V_L$ of the random potential corresponding to the spatial scale, $R_L$, can be found from the relation Eq. (4)

$$V_L R_L^3 = \gamma.$$  

(5)

An estimate for the width of the exciton level comes from the uncertainty principle,

$$\left(\frac{\hbar^2}{MV_L}\right)^{1/2} = R_L,$$  

(6)

stating that the de-Broglie wavelength corresponding to the energy $V_L$ is of the order of $R_L$. Solving the system Eqs. (5), (6) we find

$$R_L = \frac{\hbar^4}{M^2 \gamma}.$$  

(7)

We can now go back to the above assumption $R_L \gg a$. Using Eq. (7), this assumption can be cast in the form of the condition

$$\gamma \ll \frac{\hbar^4}{M^2 a}.$$  

(8)

If this condition is met, the magnitude, $V_L$, should be identified with the width of the exciton line

$$\Gamma \sim V_L = \frac{\gamma^2 M^3}{\hbar^6}.$$  

(9)

For the above picture to be consistent, one should ensure that the exciton is not destroyed by the disorder. This implies that the width, $\Gamma$, is much smaller than the Bohr energy $E_B = \frac{\hbar^2}{2a^2}$. From Eq. (9) the ratio $\Gamma/E_B$ can be expressed as

$$\frac{\Gamma}{E_B} = \frac{\mu}{M} \left(\frac{\gamma M^2 a^2}{\hbar^4}\right)^2.$$  

(10)

Comparing Eqs. (8) and (10), we conclude that the conditions $a \ll R_L$ and $\Gamma \ll E_B$ are equivalent to each other. Overall scenario of the disorder-induced broadening of the exciton line is illustrated in Fig. 1. The exciton, as an entity, is trapped in a shallow fluctuation-induced potential well of a large size. The depth of the potential well is the exciton linewidth. Within this scenario, the linewidth is independent of the Bohr radius.

The above reasoning was introduced in Refs. [15][17]. In Ref. [17] a numerical factor in the expression Eq. (9) was estimated to be 0.08.

### III. DISORDER-INDUCED SHIFT

While the long-range potential fluctuations with characteristic scale $R_L \gg a$ are responsible for inhomogeneous broadening of the exciton line, the short-range fluctuations with a scale $\sim a$ lead to another observable effect: disorder-induced shift of the exciton levels. See Fig. 1. We will illustrate this effect for the ground state of the exciton with a wave function

$$\psi_{1S}(r) = \frac{1}{\pi^{1/2} a^{3/2}} \exp \left(-\frac{r}{a}\right).$$

(11)

For this purpose, we will specify the form of the effective random potential acting on the center of mass.

Searching for the solution of the Schrödinger equation

$$\hat{H} \Psi(R, r) = E \Psi(R, r)$$

in the form

$$\Psi(R, r) = C_{1S}(R) \psi_{1S}(r).$$

(12)

Smallness of the effect of disorder allows one to neglect the admixture of the higher states of the exciton to the wave function and obtain a closed equation for $C_{1S}(R)$

$$-\frac{\hbar^2}{2M} \Delta R C_{1S}(R) + V_{eff}(R) C_{1S}(R) = (E - E_B) C_{1S}(R),$$

(13)

where the effective potential $V_{eff}(R)$ is defined as

$$V_{eff}(R) = \int dr \ V\left(R + \frac{m_h}{M} r\right) \left(\psi_{1S}(r)\right)^2.$$  

(14)

The structure of the effective potential suggests that the value of $V_{eff}$ at some point, $R$, is the result of interaction of electron with the disorder potential in the course of orbiting around the center of mass located at $R$.

Correlation properties of the effective potential follow from the relation Eq. (2) for the bare potential...
$\langle V_{eff}(\mathbf{R}_1)V_{eff}(\mathbf{R}_2) \rangle = \int d\mathbf{r}_1 d\mathbf{r}_2 \langle V(\mathbf{R}_1 + \frac{m_h}{M} \mathbf{r}_1)V(\mathbf{R}_2 + \frac{m_h}{M} \mathbf{r}_2) \rangle \psi^2_{1S}(\mathbf{r}_1)\psi^2_{1S}(\mathbf{r}_2)$
$= \gamma \int d\mathbf{r}_1 d\mathbf{r}_2 \delta(\mathbf{R}_1 + \frac{m_h}{M} \mathbf{r}_1 - \mathbf{R}_2 + \frac{m_h}{M} \mathbf{r}_2) \psi^2_{1S}(\mathbf{r}_1)\psi^2_{1S}(\mathbf{r}_2). \quad (15)$

It is now convenient to switch to the Fourier transform of the $\delta$-function, which allows to reduce the number of integrations in Eq. (15). One has

$\langle V_{eff}(\mathbf{R}_1)V_{eff}(\mathbf{R}_2) \rangle = \frac{\gamma}{(2\pi)^3} \int d\mathbf{k} \exp \left\{ i\mathbf{k}(\mathbf{R}_1 - \mathbf{R}_2) \right\}$
$\times \int d\mathbf{r} \psi_{1S}(\mathbf{r})^2 \exp \left\{ i \frac{m_h}{M} \mathbf{k} \mathbf{r} \right\}^2. \quad (16)$

From the form of the correlator, we conclude that characteristic values of $|\mathbf{k}|$ are of the order of $a^{-1}$. Thus the correlator decays at distances $|\mathbf{R}_1 - \mathbf{R}_2| > a$.

Using Eq. (16) we can express the second-order correction to the energy of the exciton moving with momentum $\mathbf{p}$ caused by $V_{eff}(\mathbf{R})$

$\delta E_p = \sum_{\mathbf{p}'} \left| \langle V_{eff}(\mathbf{R}) \rangle_{\mathbf{p},\mathbf{p}'} \right|^2 \frac{E_{\mathbf{p}'} - E_p}{E_{\mathbf{p}'} - E_p'}, \quad (17)$

where $\langle V_{eff}(\mathbf{R}) \rangle_{\mathbf{p},\mathbf{p}'}$ is the matrix element of the effective potential between the states $\mathbf{p}$ and $\mathbf{p}'$, while $E_p$ is the dispersion law of the exciton

$E(\mathbf{p}) = -E_B + \frac{\hbar^2 \mathbf{p}^2}{2M}. \quad (18)$

The pole at $E_{\mathbf{p}'} = E_p$ is responsible for the broadening of the exciton line. In order to recover the result Eq. (9) within a numerical factor, we note that the estimate for the left-hand side is $\Gamma$, while the pole in the right-hand side leads to the imaginary part proportional to the density of states at energy $E_p^{1/2} \sim \Gamma$. Since the density of the free exciton states is proportional to $E_p^{1/2}$, the right-hand side is proportional to $\Gamma^{1/2}$. Then Eq. (17) can be viewed as an equation for $\Gamma$ and yields the solution Eq. (9).

It is important to note that the imaginary part of the sum Eq. (17) comes from the momenta $p \sim R^{-1}$, while the real part is accumulated from much bigger domain of momenta $p \sim a^{-1}$. From this we conclude that the real part of the sum is a self-averaging quantity, which permits the replacement of $\left| \langle V_{eff}(\mathbf{R}) \rangle_{\mathbf{p},\mathbf{p}'} \right|^2$ by its average and neglect $E_p$ compared to $E_{\mathbf{p}'}$ in the denominator of Eq. (17). As a result, Eq. (17) turns into a down-shift of the ground-state level of the exciton

$W_{1S} = -\frac{\gamma}{(2\pi)^3} \int d\mathbf{p}' \int d\mathbf{r} \psi_{1S}(\mathbf{r})^2 \exp \left\{ -i \frac{m_h}{M} \mathbf{p}' \mathbf{r} \right\}^2 \frac{1}{E_{\mathbf{p}'} - E_p}. \quad (19)$

The remaining task is to evaluate the integrals in Eq. (19), which can be done analytically. The spatial integral is taken using the relation

$\int d\mathbf{r} \psi_{1S}(\mathbf{r}) \exp(i\mathbf{q}\mathbf{r}) = \frac{1}{\left(1 + \frac{q^2 a^2}{4} \right)^2}. \quad (20)$

Now the integral over $\mathbf{p}'$ can be made dimensionless and evaluated. The result reads

$W_{1S} = -\frac{\gamma M^2}{\pi^2 \hbar^2 m a^2} \int_0^\infty \frac{dz}{\left(1 + \frac{z^2}{4} \right)^2} = -\frac{5\gamma M^2}{16\pi \hbar^2 m a}. \quad (21)$

Recall now, that we have restricted the calculation of the shift $W_{1S}$ to the lowest dispersion branch Eq. (18) of the exciton. Neglecting the higher branches requires that $W_{1S} \ll E_B$. To check this requirement, we use Eq. (4) to present the shift in the form

$|W_{1S}| = \left( \frac{\Gamma E_B}{2} \right)^{1/2}. \quad (22)$

The above relation suggests that, when disorder does not destroy the exciton, i.e. when $\Gamma \ll E_B$, we have $\Gamma \ll W_{1S} \ll E_B$. This, in turn, suggests that the shift is the dominant effect induced by the disorder. Still, calculating this shift within the continuum of the ground-state branch is justified.

**IV. HIGHER S-STATES OF AN EXCITON**

Since the width of the 1S state is independent of the form of the wave function, it is obvious that the width of the 2S state is also equal to $\Gamma$. On the other hand, the correlator of the effective potential Eq. (16) acting on the center of mass depends on the form of $\psi_{1S}(\mathbf{r})$. Since the function $\psi_{1S}(\mathbf{r})$ is more compact than $\psi_{2S}(\mathbf{r})$, we expect that the shift for the 2S state is smaller than for the 1S state. Calculation of this shift is similar to the calculation of $W_{1S}$. The wave function of the 2S state has the form

$\psi_{2S}(\mathbf{r}) = \frac{1}{(2\pi)^{1/2} a^{3/2}} \left(2 - \frac{r}{a}\right) \exp \left(-\frac{r}{2a}\right). \quad (23)$

Calculation of the Fourier transform of $\psi_{2S}(\mathbf{r})$ is straightforward. The result reads

$\int d\mathbf{r} \psi_{2S}^2(\mathbf{r}) \exp(i\mathbf{q}\mathbf{r}) = \left(1 - q^2 a^2\right)^2 \left(1 - 2q^2 a^2\right) \left(1 + q^2 a^2\right)^4. \quad (24)$
Substituting this result into the expression Eq. (19) for the shift and performing integration over \( p' \) using Mathematica, we present the shift of the 2S state in the form

\[
W_{2S} = -D_{2S} \frac{\gamma M^2}{\pi^2 h^2 m_n a},
\]

where the constant \( D_{2S} \) is equal to

\[
D_{2S} = \frac{77\pi}{1024}.
\]

This is smaller than the corresponding value for 1S, which is \( D_{1S} = \frac{3\pi}{16} \). We calculated the value of \( D_{3S} \) using Mathematica both for the Fourier transform and for integration over \( p' \) and obtained the value

\[
D_{3S} = \frac{17\pi}{512}.
\]

For large \( n \), the dimensionless shift, \( D_{ns} \), can be calculated semiclassically. The form of the semiclassical wave function is the following

\[
\psi_{nS}(r) = A_n \frac{\sin \left[ \left( \frac{2\pi}{\kappa} \right)^{1/2} \int_0^r dr' \left( \frac{e^2}{\kappa r^2} - |E_n| \right)^{1/2} \right]}{\left( \frac{e^2}{\kappa r^2} - |E_n| \right)^{1/4}},
\]

where \( |E_n| = E_B/n^2 \) and \( A_n \) is the normalization constant. In determining \( A_n \), the sine square in \( \psi_{nS}(r)^2 \) can be replaced by \( \frac{1}{2} \). Equally, this replacement is justified in calculation of the Fourier transform

\[
\int dr \, \psi_{nS}(r)^2 \exp(irq) = 2\pi \int_0^{r_n} dr \frac{A_n^2}{\left( \frac{e^2}{\kappa r^2} - |E_n| \right)^{1/2}} \left( \frac{2 \sin qr}{qr} \right),
\]

where \( r_n = a_B n^2 \) is the radius on \( n \)-th orbit. The reason why the oscillations in \( \psi_{nS}^2 \) can be averaged out is that the characteristic \( q \) in Eq. (29) is of the order of \( 1/r_n \), while \( \psi_{nS} \) oscillates \( n \gg 1 \) times between \( r = 0 \) and \( r = r_n \).

Integral Eq. (29) can be evaluated explicitly upon substitution of \( r = \frac{1}{2} r_n (1 - y) \), after which it transforms as follows

\[
\int dr \, \psi_{nS}(r)^2 \exp(irq) = \frac{2\pi A_n^2}{q |E_n|^{1/2}} \int_0^1 dy \frac{\sin \left[ \frac{2\pi}{2} (1 - y) \right]}{(1 - y^2)^{1/2}}.
\]

Notice, that only an even in \( y \) term in the numerator of Eq. (30) contributes to the integral. It then reduces to the zero-order Bessel function. The final result reads

\[
\int dr \, \psi_{nS}(r)^2 \exp(irq) = \frac{\sin \left( \frac{2\pi}{2} \right)}{\left( \frac{2\pi}{2} \right)} J_0 \left( \frac{qr_n}{2} \right).
\]

We see that the right-hand-side turns to 1 in the limit \( q \to 0 \), as it should be. We also see that characteristic \( q \) is indeed \( \frac{1}{r_n} \). This leads to the dependence \( D_{nS} \propto \frac{1}{r_n} \propto \frac{1}{n^2} \). With numerical factor specified, we get

\[
D_{nS} = \frac{2}{n^2} \int_0^\infty dx \left( \frac{\sin x}{x} \right)^2 J_0^2(x) = \frac{1.86}{n^2}.
\]

Note, however, that increase of \( n \) is limited by the condition \( R_L \gg r_n \).

V. 2P AND 3P-STATES

Within the simplest model of the exciton adopted in the present paper, only the S-states are optically active. Still, P-states of the exciton can be accessed if the absorption is assisted by the disorder. Disorder-induced broadening of S and P-states are the same, but the disorder-induced shifts of the P-states are different. It is not obvious how the shifts of the levels with the same \( n \) are related to each other. To find out, we calculate the shift for the 2P-states.

We start from the wave function of the 2P-state

\[
\psi_{2P}(r) = \frac{1}{4(2\pi)^{1/2} a^{3/2}} \left( \frac{r}{a} \right) \exp \left( -\frac{r}{2a} \right) \cos \theta,
\]

where \( \theta \) is a polar angle. Calculation of the Fourier transform is straightforward and yields

\[
\int dr \, \psi_{2P}^2 \exp(iqr) = \left( 1 - 5q^2a^2 \right) \left( 1 + q^2a^2 \right)^{-1}.
\]

The shift is then again calculated using Mathematica. Casting the result in the form Eq. (25), we find

\[
D_{2P} = \frac{81\pi}{1024}.
\]

The fact that \( D_{2P} \) exceeds \( D_{2S} \) given by Eq. (26) is our main result. Our calculation shows that the first excited state, being degenerate in the absence of disorder, is split by the disorder. Renormalized 2P-level lies below the renormalized 2S-level. Although the relative splitting is small \( \approx 4\% \), it exceeds parametrically the disorder-induced broadening.

We have also checked that the shifts 3S and 3P levels are close to each other. Namely, for \( D_{3P} \) we obtained the value

\[
D_{3P} = \frac{199\pi}{6144},
\]

which is only 2.5% bigger than \( D_{3S} \) given by Eq. (27).

VI. CONCLUDING REMARKS

(i) Behavior of dimensionless shift, \( D_{nS} \propto \frac{1}{r_n} \), can be interpreted as disorder-induced renormalization of the
Bohr energy:

$$E_B = E_B \left(1 + \frac{1.85\gamma M^2}{\pi^2\hbar^2 m_e a E_B} \right).$$  \hfill (37)

To perform an estimate of the correction, we use Eq. (22). Within a numerical factor, the relative correction can be cast in the form \( (\Gamma/E_B)^{1/2} \), so that the value of the width and of the Bohr energy can be taken from experiment.\(^\text{[5]}\) Choosing the values \( \Gamma = 0.1\text{meV} \) and \( E_B = 100\text{meV} \), we get the value 3\% for the relative correction.

(ii) When calculating the disorder-induced splitting of the 2S and 2P states we assumed that short-range fluctuations scatter 2S states into 2S states, and the same fluctuations scatter 2P states into 2P states. We neglected the contribution to the shift from 2S \( \leftrightarrow \) 2P processes. These processes can be taken into account explicitly if \( \psi_{1S}(\mathbf{r})^2 \) in Eq. 19 is replaced by the product \( \psi_{2S}(\mathbf{r})\psi_{2P}(\mathbf{r}) \). The Fourier transform of this product is given by

$$\int d\mathbf{r}\psi_{2S}(\mathbf{r})\psi_{2P}(\mathbf{r}) \exp(iq\mathbf{r}) = \frac{3qa(1-q^2a^2)}{(1+q^2a^2)^2}. \hfill (38)$$

Calculating the shift from this Fourier transform yields a contribution to \( D_{2S} \) equal to \( \frac{4\pi^2}{1004} \), which is comparable to the contribution \( \frac{777}{1024} \) prior. However, it is important to note that 2S \( \leftrightarrow \) 2P processes do not contribute to the splitting. This is because the constant \( \frac{777}{1024} \) adds to \( D_{2P} \) as well. Thus, the 2S \( \leftrightarrow \) 2P contributions cancel out from the splitting.

More formally, joint renormalizations of S and P exciton branches emerges as diagonal matrix elements of the self-energy matrix \( \int d\mathbf{r}(\tilde{G}\mathbf{q} U) \), where \( \tilde{G} \) is a bare \( 2 \times 2 \) Green function, while \( U(\mathbf{r}) \) is a perturbation matrix defined as

$$\tilde{U}_{ij} = \int d\mathbf{r}\psi_i(\mathbf{r})\psi_j(\mathbf{r})V(\mathbf{R} + \frac{m_e}{M}\mathbf{r}), \hfill (39)$$

where subindices \( (i, j) \) assume the values S and P.

(iii) To put the disorder-induced splitting of 2S and 2P levels of the exciton into the perspective, it is instructive to invoke the Lamb shift\(^\text{[27–29]}\) in quantum electrodynamics. In a celebrated experiment\(^\text{[24]}\) 2S and 2P hydrogen levels were found to be split by \( \approx 4.37\text{meV} \), a quantity that can be accounted for by the electron interaction with vacuum fluctuations. Theoretically, the shift of a given level is proportional to \( \psi^2(0) \), which is the probability density to find electron in the state, \( \psi(\mathbf{r}) \), at the origin\(^\text{[28]}\). As a result, the 2P state with \( \psi(0) = 0 \) remains unshifted, while the 2S state lies above the 2P state. By contrast, for an exciton in a fluctuating field of randomly positioned impurities, both 2S and 2P levels are shifted, and the 2P level is shifted down more than 2S.

(iv) It is well known\(^\text{[21,22]}\) that the white-noise disorder leads to the diverging self-energy of the electron which is cut off only by a finite correlation radius of the disorder. For exciton, the cutoff is provided by a finite Bohr radius, \( a \). Our prime observation is that interaction of 2S and 2P states with the disorder are slightly different.

(v) In transition metal dichalcogenides, see e.g. Ref. 23 and the review Ref. 24 a sequence of excited states of exciton in atomically thin samples was identified. Still, accidental degeneracy of the exciton states in dichalcogenides is missing. This is because the attraction of an electron and a hole deviates from purely Coulomb interaction at short distances\(^\text{[25]}\).

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