Tunable Excitons in Biased Bilayer Graphene

Cheol-Hwan Park and Steven G. Louie

Department of Physics, University of California, Berkeley, California 94720 USA
Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720 USA

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Recent measurements have shown that a continuously tunable bandgap of up to 250 meV can be generated in biased bilayer graphene [Y. Zhang et al., Nature 459, 820 (2009)], opening up a pathway for possible graphene-based nanoelectronic and nanophotonic devices operating at room temperature. Here, we show that the optical response of this system is dominated by bound excitons. The main feature of the optical absorbance spectrum is determined by a single symmetric peak arising from excitons, a profile that is markedly different from that of an interband transition picture. Under laboratory conditions, the binding energy of the excitons may be tuned with the external bias going from zero to several tens of meV's. These novel strong excitonic behaviors result from a peculiar, effective “one-dimensional” joint density of states and a continuously-tunable bandgap in biased bilayer graphene. Moreover, we show that the electronic structure (level degeneracy, optical selection rules, etc.) of the bound excitons in a biased bilayer graphene is markedly different from that of a two-dimensional hydrogen atom because of the pseudospin physics.

The low-energy electronic states of graphene are described by a massless Dirac equation \( \frac{\hbar v_F}{m_e} \). If an extra layer is added [Fig. 1(a)], the electronic properties change drastically and the charge carriers become massive [Fig. 1(b)] [4]. There have been a number of theoretical studies on the possibility of opening up a bandgap in the gapless bilayer graphene if an electric field is applied perpendicular to it by evaluating the optical matrix elements [36] using the action kernel [36]. The absorption spectrum is calculated by evaluating the optical matrix elements [36] using the eigenstates and eigenvalues of the Bethe-Salpeter equation (BSE):

\[ (E_{ck} - E_{ck}) A_{ck} + \sum_{c'v'k'} \langle cvk | K^{eh} | c'v'k' \rangle A_{c'v'k'} = \Omega^S A_{cck}^S, \]

where \( A_{cck}^S \) is the amplitude of a free e-h pair configuration composed of the electron state \( |ck \rangle \) and the hole state \( |v \rangle \), \( \Omega^S \) is the exciton excitation energy, \( E_{ck} \) and \( E_{ck} \) are quasiparticle energies, and \( K^{eh} \) is the e-h interaction kernel [36]. The absorption spectrum is calculated by evaluating the optical matrix elements [36] using the eigenstates and eigenvalues of the BSE.

As in recent experiments [27, 41], we focus here on the case in which the net charge on the BBG is zero, or, the displacement fields \( D \) above and below the bilayer graphene are the same [Fig. 1(c)]. We find that the optical response of BBG is dominated by low-energy bound excitons with huge oscillator strength due to the 1D nature in the joint density of states. As a consequence, the main peak of the absorbance profile becomes highly symmetric. The binding energy and oscillator strength of the excitons increase with the bandgap. We find a very rich electronic structure for the excitons in a BBG. Especially, we discover a symmetry breaking of excitons having angular momenta of equal magnitude but opposite sign which leads to an unusual selection rule in the optical absorption. This phenomenon is explained in terms of...
the pseudospin, a degree of freedom describing the bonding character between neighboring carbon atoms \([3]\), in a BBG.

In this study, we make use of the \(k\cdot p\) based method developed by Ando and coworkers for the excitonic spectra of graphene and carbon nanotubes \([42, 46]\). Although, unlike the first-principles GW-BSE approach \([39]\) that is parameter free, the current method is based on a tight-binding formalism and treats electron-electron interactions within the screened Hartree-Fock approximation, it does provide excitonic features of the absorption profile that may be compared with experiments for complex structures and applied fields \([44, 47]\). For the \(e\cdot h\) kernel \(K^{eh}\), we consider only the attractive direct term, which is by far dominant and describes the screened interaction between electrons and holes, and neglect the repulsive exchange term. The exchange kernel is responsible for singlet-triplet splitting and the splitting among states within individual singlet and triplet complexes, but is usually only a few percent in magnitude of the direct term \([37]\).

The quasiparticle energies \(E_{ck} = \varepsilon_{ck} + \Sigma_{ck}\) and \(E_{ek} = \varepsilon_{ek} + \Sigma_{ek}\) are obtained by first calculating the bare energy \(\varepsilon_k\) within the \(k\cdot p\) formalism \([44]\) using a tight-binding Hamiltonian where we set the intralayer hopping parameter between the nearest-neighboring atoms \(\gamma_0 = 2.6\) eV and the interlayer hopping parameter \(\gamma_1 = 0.37\) eV. These parameters reproduce well the bandstructure of pristine bilayer graphene obtained from density-functional calculations within the local density approximation (LDA) \([30]\). The self energy \(\Sigma_k\) is calculated within the screened Hartree-Fock approximation, using the static random-phase dielectric function \([42–44, 47]\). We calculate the static polarizability within the random-phase approximation \([42, 44, 47]\) by including the four electronic bands closest to the bandgap arising from the \(\pi\) states with an energy cutoff of 5 eV (we have checked that the resulting quasiparticle energies are insensitive to this cutoff), and incorporate the effects of screening from higher-energy states (including the \(\pi\) bands away from the Dirac points and the \(\sigma\) bands) by an additional effective static dielectric constant \(\epsilon_{\text{int}} = 2.0\) as done in previous graphene and nanotube studies \([45, 47]\). The total dielectric function \(\epsilon(q)\) is given by \(\epsilon(q) = 1 - v(q) \left[ P_{\text{int}}(q) + P(q) \right]\) where \(v(q) = 2\pi\epsilon^2/q\) is the bare Coulomb interaction and \(P_{\text{int}}(q)\) and \(P(q)\) are the static polarizabilities coming from \(e\cdot h\) excitations involving higher-energy states and those involving only the low-energy \(\pi\) states, respectively. Using the relation \(\epsilon_{\text{int}}(q) = 1 - v(q) \left[ P_{\text{int}}(q) \approx \epsilon_{\text{int}} \right.\) for screening with low-momentum transfer, we obtain \(\epsilon(q) \approx \epsilon_{\text{int}} - v(q) \left. P(q) \right|_{q < 3}\). The calculated self energy is then added to the LDA band energy to form the quasiparticle energy. Although in this scheme, the LDA exchange-correlation energy is not subtracted from the LDA band energy, it should be a reasonable approximation because the LDA exchange-correlation energy is nearly the same for all the \(\pi\) states giving rise to a constant shift to both occupied and unoccupied states.

We use in all the calculations a very dense grid for electronic state sampling corresponding to \(1500 \times 1500\) \(k\)-points in the irreducible wedge of the Brillouin zone of bilayer graphene in order to describe the extended wavefunction \((e\cdot h\) correlation length\) of the excitons in real space, in particular at small bias voltage when the bandgap is small.

The external displacement field \(D\) induces an imbalance between the charge densities on the two graphene layers of the BBG, which creates an internal depolarization electric field. This depolarization field induces additional charge changes, which in turn induce further adjustments in the internal electric field, and so on. We obtain the resulting internal electric field and the charge imbalance between the layers by solving Poisson’s equation \([27]\).

Figure 1(e) is a schematic diagram showing the squared \(e\cdot h\) amplitude (wavefunction) of the lowest-energy optically active (bright) exciton for incident light with in-plane polarization. |\(\Psi(r_e, r_h = 0)|^2| = \sum_{c,v} A_{c,v}^2 \langle r_e | c,k \rangle \langle v | k | r_h = 0 \rangle|^2|\) where the hole is fixed at a carbon atom belonging to the \(B’\) sublattice. The bound excitons [Fig. 2(a)] are comprised of interband transitions forming the bandgap [Fig. 1(d)]. The electronic states in those two bands are localized at \(A\) and \(B’\) sublattices for the conduction and valence band, respectively, i.e., the electron and hole are localized on two different graphene layers. As shown schematically in Fig. 1(e), the position of the maximum in the electron density is not on top of the hole, but is on a ring with a radius \(R_{eh}\) from the hole that is about two orders of magnitude larger than the interlayer distance \(d\). The radius of the exciton in real space is related to that in \(k\) space by \(R_{eh} \approx 2\pi / R_{ck}\) [see, e.g., Fig. 2(e) and Fig. 2(f)].

Figure 2(a) shows the bound exciton levels for a particular BBG \((eDd = 0.40\) eV\), in which we label an exciton by the radial quantum number \(n\) and the angular momentum quantum number \(m\) of its wavefunction. The wavefunction of an exciton \(X_{n,m}\) formed from the free \(e\cdot h\) pairs near the \(K\) point is approximately of the form \(\Psi(r_e, r_h = 0) \approx e^{im\theta_{r_e}} e^{i|m|f_{n,m}(r_e)}\) near the origin \((r_e = 0)\) where \(f_{n,m}(r_e)\) has \(n\) zeros like the wavefunctions in the 2D quantum well problem having the angular symmetry \([48]\). However, in our system, the angular symmetry is broken, i.e., the binding energy of \(X_{n,m}\) and that of \(X_{n,−m}\) are different (see Table 1). The origin of this symmetry breaking lies in the pseudospin of BBG. The electronic states in in the conduction and valence bands forming the bandgap of a BBG, in the basis of amplitudes on the four sublattices \((A, B, A’\) and \(B’\), are

\[
|m| = (a_1, a_2 e^{i\theta_k}, a_3 e^{i\theta_k}, a_4 e^{2i\theta_k})^T
\]
TABLE I: Calculated quantities of bound excitons in a BBG: the binding energy ($E_b$), the radial quantum number $n$, the angular momentum quantum number $m$, and the integrated absorbance (IA), the absorbance integrated over energy, of the exciton $X_{n,m}$ made from free $e$-$h$ pairs near the K point. The IA is for incident light with in-plane polarization. The quantities are the same for the exciton $X'_{n,-m}$ made from $e$-$h$ pairs near the K' point. Here, we consider the BBG with $V_{cat} = eDd$ equal to 0.40 eV.

| Index | $E_b$ (meV) | $n$ | $m$ | $n + |m|$ | IA (meV) |
|-------|-------------|-----|-----|----------|----------|
| 1     | 55.6        | 0   | 0   | 0        | 0.000    |
| (bright) 2 | 40.6   | 0   | -1  | 1        | 1.240    |
| 3     | 35.0        | 0   | 1   | 1        | 0.000    |
| 4     | 32.7        | 0   | -2  | 2        | 0.000    |
| 5     | 27.0        | 0   | 2   | 2        | 0.000    |
| 6     | 25.5        | 0   | -3  | 3        | 0.000    |
| 7     | 22.8        | 1   | 0   | 1        | 0.000    |
| 8     | 22.0        | 0   | 3   | 3        | 0.000    |
| 9     | 20.9        | 0   | -4  | 4        | 0.000    |
| (bright) 10 | 19.5   | 1   | -1  | 2        | 0.146    |
| 11    | 18.5        | 0   | 4   | 4        | 0.000    |
| 12    | 18.2        | 1   | 1   | 2        | 0.000    |
| 13    | 17.7        | 0   | -5  | 5        | 0.000    |
| 14    | 17.0        | 1   | -2  | 3        | 0.000    |
| 15    | 15.9        | 0   | 5   | 5        | 0.000    |
| 16    | 15.4        | 1   | 2   | 3        | 0.000    |
| 17    | 15.2        | 0   | -6  | 6        | 0.000    |
| 18    | 14.7        | 1   | -3  | 4        | 0.000    |
| 19    | 14.4        | 2   | 0   | 2        | 0.000    |
| 20    | 13.9        | 0   | 6   | 6        | 0.000    |
| 21    | 13.3        | 0   | -7  | 7        | 0.000    |
| 22    | 13.2        | 1   | 3   | 4        | 0.000    |
| 23    | 12.7        | 1   | -4  | 5        | 0.000    |
| (bright) 24 | 12.4   | 2   | -1  | 3        | 0.093    |
| 25    | 12.3        | 0   | 7   | 7        | 0.000    |

and

$$|v\mathbf{k}\rangle \propto (-a_4, a_3 e^{i\theta}, -a_2 e^{i\theta}, a_1 e^{2i\theta})^T,$$  

(3)

respectively, where $a_i$'s ($i = 1, 2, 3, 4$) are real constants. As discussed above, the band edge states that form the bound excitons have $|a_1| \approx 1$ and $|a_2|, |a_3|, |a_4| \ll 1$, i.e., the electron and hole are localized at the A and B' sublattices, respectively. Therefore, the pseudospin of the states in a BBG imposes approximately an extra phase of $e^{-2i\theta}$ to the $e$-$h$ pair state $|v\mathbf{k}\rangle\langle c\mathbf{k}|$, resulting in an extra pseudospin angular momentum quantum number $m_{ps} = -2$. This behavior is unique in BBG. In pristine bilayer graphene, $|a_1|$ and $|a_4|$ are the same [43, 44], and hence we cannot define a single extra phase.

If we denote the angular momentum of an exciton coming from the envelope function $A^S_{cv\mathbf{k}}$ by $m_{env}$, then the total angular momentum quantum number (which is the approximate good quantum number) is given by $m = m_{env} + m_{ps}$. Because of the extra pseudospin angular momentum, two exciton states having $m_{env}$ of the same magnitude but of opposite sign are no longer degenerate since $m$ would be different. Rather, two states having total angular momentum quantum number $m$ and $-m$ will be degenerate if the extra phase imposition by the pseudospin is perfect. In fact, the extra phase imposition of $e^{-2i\theta}$ is not perfect because the coefficients $|a_2|, |a_3|$, and $|a_4|$ are non-zero, resulting in the degeneracy breaking shown in Table I. [The broken angular symmetry shown, e.g., in Fig. 2(m) has the same origin.] On the contrary, due to time-reversal symmetry, the exciton $X_{n,m}$ (formed by states near K) is degenerate in binding energy with $X'_{n,-m}$, which is an exciton made from the free $e$-$h$ pairs near the K' point with radial and angular momentum quantum numbers $n$ and $-m$, respectively. Therefore, considering the spin and valley degeneracy and neglecting possible intervalley coupling, each bound exciton shown in Fig. 2(a) is four-fold degenerate.

The extra phase $e^{-2i\theta}$ arising from the pseudospin in a BBG qualitatively changes the selection rule for optical absorption as follows. The oscillator strength $O$ of an exciton $S$ of a BBG is given by $O = \sum_{cv\mathbf{k}} A^S_{cv\mathbf{k}} \langle v\mathbf{k}|\hat{O}|c\mathbf{k}\rangle$ in which $\hat{O}$ is proportional to the electron-photon interaction Hamiltonian. If the exciting photons are polarized along the $z$ direction (i.e., parallel to the graphene planes), then $\hat{O} \propto \begin{pmatrix} \sigma_x & 0 \\ 0 & -\sigma_x \end{pmatrix}$, where $\sigma_x$ is the Pauli matrix [46]. Using Eqs. (2) and (3), we obtain $O \propto \sum_{cv\mathbf{k}} A^S_{cv\mathbf{k}} (a_1 a_3 e^{-i\theta} - a_2 a_4 e^{i\theta})$. In order to have a non-vanishing oscillator strength, we should have $A^S_{cv\mathbf{k}} \propto e^{i\theta}$ or $A^S_{cv\mathbf{k}} \propto e^{-i\theta}$, i.e., the envelope angular momentum quantum number $m_{env}$ should be 1 or $-1$. Therefore, the total angular momentum quantum number $m$ (which is equal to $m_{env} - 2$) for the optically active excitons is either $-1$ or $-3$. However, since $|a_1|$ is by far the largest among the four $|a_i|$'s and $|a_1 a_3| \gg |a_2 a_4|$ [43], effectively, only the excitons $X_{n,-1}$ or $X'_{n,1}$ are optically active (Table I). This unusual optical selection rule in a BBG, hence, originates from the unique pseudospin physics.

In the discussion below on the optical absorbance, for concreteness, we shall assume that the polarization of the incident light is linear and is parallel to the graphene planes. Accordingly, the lowest-energy exciton $X_{0,0}$ [Figs. 2(b), 2(d)] is dark and the second lowest-energy exciton $X_{0,-1}$ [Figs. 2(e), 2(g)] is bright. As seen from the calculated oscillator strength in Table I, the lowest-energy bright excitons by far dominate the absorbance spectrum. The first, second and third bright excitons have zero, one, and two nodes in the exciton wavefunction along the radial direction, respectively, in both momentum and real space (Fig. 2). Also, there are many dark exciton levels between the bright exciton ones as
shown in Fig. 2(a). A change in the polarization direction of the incident light away from the graphene plane would alter the optical strength of the levels from those given in Fig. 2(a).

In a 2D hydrogen atom, the binding energy is proportional to $(n + |m| + 1/2)^{-2}$, resulting in a $2N + 1$-fold degeneracy with $N = n + |m|$. As shown in Table I, however, this degeneracy in the binding energy of the excitons in a BBG is broken, and, further, the order of the binding energies largely deviates from the case for a 2D hydrogenic model. Also, we have checked that the detailed order of exciton levels changes with the external displacement field.

Figure 3 shows the calculated absorbance spectrum of BBG (for in-plane linearly polarized incident light) near the bandgap energy and the wavefunction of the lowest-energy bright exciton that forms the main peak for several bias voltages. Remarkably, when $e$-$h$ interactions are accounted for, the absorbance profile is dominated by a single four-fold degenerate excitonic level with huge oscillator strength. Accordingly, the dominant feature of the absorbance profile near the bandgap energy becomes symmetric when excitonic effects are considered – as in carbon nanotubes [37, 49]; whereas, if these effects are neglected, highly asymmetric absorbance spectra are obtained reflecting the “effective” 1D van Hove singularity in the joint density of quasiparticle states discussed above. The huge excitonic effects observed here in fact originate from this 1D singularity [37] which becomes more and more dominant as the bandgap increases. On the contrary, excitonic effects on the low-energy ($\leq 1$ eV) optical response of pristine bilayer graphene are negligible since its joint density of states is characteristic of a 2D system [30]. The enhancement of excitonic effects with the bandgap is reflected in the increase in the exciton binding energy [Fig. 3(a)] and the decrease in the exciton radius [Fig. 3 and Fig. 3(b)].

In a previous study [41], we have shown that when the photo-excitation energy is close to the energy of the zone-center optical phonons in BBG ($\sim 0.2$ eV), Fano line-shapes in the absorbance profile develop due to the coupling of $e$-$h$ pair excitations with the phonons. We expect that similar exciton-phonon coupling behavior, whose effects on the optical response is large when the optical energy gap is around 0.2 eV, would arise if electron-phonon interactions are taken into account.

The above results are applicable to suspended BBG [50]. However, for BBG on substrates, excitonic effects are altered due to enhanced screening from the substrate. As an example, we consider the effect of background screening due to the substrate on the optical response of BBG relevant for the experimental setup in Refs. 27 and [41]. For substrates above and below the BBG having dielectric constants $\epsilon_1$ and $\epsilon_2$, respectively, their effect can effectively be replaced by a single material having a dielectric constant of $\epsilon_{BG} = (\epsilon_1 + \epsilon_2)/2$ [51].

Using the static dielectric constant of SiO$_2$ (=3.9) and that of amorphous Al$_2$O$_3$ (=7.5), we may roughly set the external background dielectric screening as $\epsilon_{BG} = (3.9 + 7.5)/2 = 5.7$. Figure 3 shows similarly calculated quantities as in Fig. 3(a), but now for BBG with added substrate screening as discussed above. The exciton binding energy $E_b = \Delta^{QP} - \Delta^{BSE}$ is smaller than the case without substrate screening. The calculated optical gap $\Delta^{BSE}$ is in reasonable agreement with the experiment $\Delta^{Exp}$ [27].

In this work, the inter-layer trigonal warping effects on the electronic structure (owing to an atom on one layer interacting with further neighbors on the other layer), i.e., the trigonal anisotropy in the energy band dispersion near a Dirac point [4], have been neglected. If these effects were taken into account, the cylindrical symmetry about an individual Dirac point is weakly broken, leading to only minor changes in exciton energies and to some of the dark excitons gaining very small optical oscillator strength. However, the change in the overall absorbance spectra (which are dominated by excitons) at different gaps is negligible [52].

In conclusion, we have shown that excitons in biased bilayer graphene dramatically change the optical response because of the 1D nature of the joint density of quasiparticle states in this system. These excitonic effects are remarkably tunable by the external electric field. Also, we have shown that the pseudospin character of the electronic states dramatically alters the excitonic structure (energy level degeneracy, optical selection rule, etc.) of this system. These results illustrate the richness in the photophysics of biased bilayer graphene and their promise for potential applications in nanoelectronic and nanophotonic devices at room temperature [27].

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* Electronic address: sglouie@berkeley.edu

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FIG. 1: (a) Schematic diagram showing the structure of pristine bilayer graphene whose unit cell is composed of four different sublattices (A, B, A’, and B’). (b) Schematic bandstructure of pristine bilayer graphene (origin is the Dirac point). Solid blue and dashed red lines represent valence bands and conduction bands, respectively. (c) and (d): Same schematic diagrams as in (a) and (b) for bilayer graphene under a displacement field D generated through a double-gate. In (d), Δ is the energy bandgap and vertical arrows represent interband transitions responsible for the formation of excitons. (e) Schematic diagram showing the probability density that a photo-excited electron is found at \( r_e \) when the hole (blue empty circle) is fixed at the origin. \( |\Phi(r_e, r_h = 0)|^2 \) (see text). For visualization purposes, we show the quantities in a vertical plane that includes the hole. The fake thickness of the plotted profile (red) is proportional to the probability density. The interlayer distance \( d \) is extremely exaggerated in (e). The size of the exciton \( R_{eh} \) is much larger than \( d \) [Fig. 1(b)].

FIG. 2: (a) Calculated free e-h pair excitation dispersion \( E_{\phi k} - E_{\phi k} \) versus \( k \) and exciton levels of a BBG with an external electrostatic potential between the two graphene layers \( V_{ext} \) equal to 0.40 eV (Fig. 1). Thick red lines and thin blue lines show optically active (bright) and inactive (dark) exciton levels, respectively, for incident light with in-plane polarization. The exciton \( X_{n,m} (X'_{n,m}) \) formed by e-h pairs near the K (K’) point is denoted by its radial quantum number \( n \), angular momentum quantum number \( m \) (see text), and binding energy \( E_b \). Each exciton level is four-fold degenerate due to the spin and valley degeneracy (see text). There are many other higher-energy bound excitons not shown here whose energy is below the bandgap. (b) The squared amplitude of the lowest-energy exciton \( \text{[exciton wavefunction in real space of the corresponding exciton in (b)]} \) in momentum space \( |A_{\phi k}^2| \). (c) Squared wavefunction in real space of the corresponding exciton in (b). The plotted quantity is the probability density \( |\Phi(r_e, r_h = 0)|^2 \) of finding an electron at \( r_e \) given that the hole is fixed at one of the carbon atoms (at the center of the figure) in sublattice B’ (Fig. 1). (d) Real part of the exciton wavefunction Re \( \Phi(r_e, r_h = 0) \) for the corresponding exciton in (b). (g)-(i), (h)-(j), and (k)-(m): Similar quantities as in (b)-(d) for the first, the second and the third bright excitons [excitons \( X_{0,1}, X_{1,1}, \) and \( X_{2,-1} \) in (a), respectively].

FIG. 3: (a) Calculated absorbance spectra of BBG (with an arbitrary energy broadening of 5 meV and in-plane polarization) where \( V_{ext} = eDd \) (see Fig. 1) is 0.14 eV. Results with (blue or solid line) and without (red or dashed line) e-h interaction effects are shown. (b) Wavefunction of the lowest-energy bright exciton \( (X_{0,1} \text{ or } X'_{0,1}) \) that forms the dominant peak in the absorbance spectrum. The plotted quantity is the probability density \( |\Phi(r_e, r_h = 0)|^2 \) of finding an electron at \( r_e \) given that the hole is fixed at one of the carbon atoms (at the center of the figure) in sublattice B’ (see Fig. 1). (c) and (d), (e) and (f), and (g) and (h): Same quantities as in (a) and (b) for \( V_{ext} = 0.27 \text{ eV}, 0.40 \text{ eV}, \) and \( 0.66 \text{ eV} \), respectively.

FIG. 4: (a) The quasiparticle bandgap \( \Delta_{QP} \), the optical bandgap \( \Delta_{BSE} \), and the binding energy \( E_b \) (= \( \Delta_{QP} - \Delta_{BSE} \)) of BBG versus \( V_{ext} = eDd \). (b) The size \( R_{eh} \), defined in Fig. 4(e), of the lowest-energy bright exciton \( (X_{0,1} \text{ or } X'_{0,1}) \) versus \( V_{ext} \). The line is a guide to the eye.

FIG. 5: The quasiparticle bandgap \( \Delta_{QP} \), the optical bandgap \( \Delta_{BSE} \), and the binding energy \( E_b \) (= \( \Delta_{QP} - \Delta_{BSE} \)) of BBG under background screening \( (\epsilon_{BG} = 5.7) \) versus \( V_{ext} = eDd \). Measured data \( \Delta_{Exp} \) are taken from Ref. [27].