Bipolaronic blockade effect in quantum dots with negative charging energy

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Abstract – We investigate single-electron transport through quantum dots with negative charging energy induced by a polaronic energy shift. For weak dot-lead tunnel couplings, we demonstrate a bipolaronic blockade effect at low biases which suppresses the oscillating linear conductance, while the conductance resonances under large biases are enhanced. A novel conductance plateau develops when the coupling asymmetry is introduced, with its height and width tuned by the coupling strength and external magnetic field. It is further shown that the amplitude ratio of the magnetic-split conductance peaks changes from 3 to 1 for increasing coupling asymmetry. Though we demonstrate all these transport phenomena in the low-order single-electron tunneling regime, they are already strikingly different from the usual Coulomb blockade physics and are easy to observe experimentally.

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Introduction. – At low temperatures, various nanostructures weakly coupled to external electrodes exhibit the Coulomb blockade of single-electron (SE) tunneling. This phenomenon has already become a classic hallmark of electronic transport through quantum dots (QDs) fabricated in semiconductor heterostructures [1], nanowires [2], carbon nanotubes [3], and even defined in single molecules or atoms [4]. Generally, SE tunneling occurs when the QD chemical potential, tuned by a gate voltage, is aligned with the transport window opening up between the Fermi energies of the source and drain. Otherwise, the transport is Coulomb-blockaded. This gives rise to diamond-shaped blockade regions surrounded by regions of SE tunneling in the differential conductance map as a function of the gate and the source-drain voltage. For vanishing source-drain voltages, the linear conductance features periodic Coulomb peaks as the gate voltage is varied. These peaks are approximatively spaced by the charging energy for adding an electron to the QD. Due to the Coulomb repulsion of electrons, the charging energy is positive, representing a key parameter for the Coulomb blockade physics in the SE tunneling regime [5]. An interesting question then arises: What will the scenario of SE tunneling be if the charging energy becomes negative?

Indeed, the possibility for inverting the sign of the charging energy has been offered by experiments on single-molecular junctions [6] and suspended carbon nanotubes [7], where the QDs are characterized by a Holstein coupling to quantized vibrational degrees of freedom. For sufficiently strong Holstein coupling, the induced polaronic shift can overcome the Coulomb repulsion, resulting in a bipolaronic attraction between electrons and thus a negative effective charging energy for the QD. This possibility has spurred some theories exploring the manifestation of negative charging energy in transport, which mainly concern the charge Kondo effect [8], pair tunneling [9–11], and cotunneling [10]. However, these high-order transport behaviors are fragile, requiring harsh experimental conditions. For example, the charge Kondo effect and pair tunneling only occur at the particle-hole symmetric point with extremely small energy scales. Any deviation from the symmetric point will suppress the Kondo correlations. The pair tunneling is also subject to

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the exponential Franck-Condon suppression and is difficult to observe from a broad background of other tunneling contributions. It is hence highly desirable to identify transport phenomena characteristic for negative charging energy in the lower-order SE tunneling regime that is easy to access experimentally.

In this paper, we demonstrate, as a counterpart to the conventional Coulomb blockade physics, a blockade effect of the low-order SE tunneling due to the bipolaronic attraction, which brings about strikingly distinct transport spectroscopy. For weak dot-lead tunnel couplings, while the conductance resonances at large biases are enhanced, the conductance at low biases, especially the oscillating linear conductance, is suppressed. Further asymmetrically tuning the coupling can merge certain enhanced and suppressed conductance peaks into a finite plateau. Its height and width are dependent on the coupling strength and external magnetic field. We also find that the magnetic field splits conductance resonances into two peaks with a novel amplitude ratio of 3 for symmetric couplings and identical part. These phenomena are explained by the unusual behavior of electron occupation on the dot along with the energy resonance conditions for SE tunneling.

**Model Hamiltonian and formulation.** – We start from modeling the QD with one localized orbital [12] coupled to a vibrational mode as well as to the left (L) and right (R) leads by the Anderson-Holstein Hamiltonian, which reads

\[ H = \sum_\sigma \varepsilon_\sigma \hat{d}_\sigma \hat{d}_\sigma^\dagger + U \hat{d}_{\sigma L} \hat{d}_{\sigma L}^\dagger \hat{d}_{\sigma R} \hat{d}_{\sigma R}^\dagger + \varepsilon \hat{b}^\dagger \hat{b} + M \hat{n} (\hat{b}^\dagger + \hat{b}) + \sum_{k,\sigma,\alpha} \varepsilon_k \hat{C}_{k\sigma\alpha}^\dagger \hat{C}_{k\sigma\alpha} + \sum_{k,\sigma,\alpha} (V_{\alpha} \hat{C}_{k\sigma\alpha}^\dagger \hat{d}_\sigma + \text{H.c.}), \]

where \( \hat{d}_\sigma^\dagger \) (\( \hat{C}_{k\sigma\alpha}^\dagger \)) creates an electron with spin \( \sigma = \uparrow, \downarrow \) and energy \( \varepsilon_\sigma \) (\( \varepsilon_k \)) in the QD orbital (in the \( \alpha \) lead, \( \alpha = L, R \), and \( U \)) denotes the on-site Coulomb repulsion. Mechanical vibrations with frequencies \( \varepsilon_p \) are excited by the phonon operator \( \hat{b}^\dagger \), which couple to the total dot charge \( \hat{n} = \sum_\sigma \hat{d}_\sigma \hat{d}_\sigma^\dagger \) through a Holstein coupling strength \( M \). Finally, electronic tunneling between the dot and leads is accounted for by tunneling matrix elements \( V_{\alpha} \) in the last term. We then apply a polaronic transformation [13] \( \hat{H} \rightarrow \tilde{H} = e^{\hat{S}} H e^{-\hat{S}} \), with \( S = (M/\varepsilon_p) \hat{n} (\hat{b}^\dagger - \hat{b}) \), to eliminate the electron-phonon coupling. As a result, the orbital energy gets renormalized \( \varepsilon_\sigma \rightarrow \varepsilon_\sigma - M^2/\varepsilon_p \), which is canceled by redefining \( \varepsilon_\sigma \). Due to the polaronic shift \( U_c \rightarrow U = U_c - 2M^2/\varepsilon_p \), the Coulomb repulsion can be renormalized downward to a bipolaronic attraction, realizing the scenario of negative charging energy. The transformation also leads to dressed tunneling matrix elements \( V_{\alpha} \rightarrow V_{\alpha} e^{(M/\varepsilon_p) (\hat{b}^\dagger - \hat{b})} \). Physically, the phonon operators arise in the tunneling term because a cloud of phonons may be created or absorbed when the electron occupation in the dot changes, due to the electron-phonon interaction. For temperatures and biases lower than the phonon frequency, vibrational excitations are energetically not allowed. In this regime, after averaging over the zero-phonon state [9,14] and redefining the dressed tunneling matrix elements as \( V_{\alpha} \), the effective Hamiltonian \( \tilde{H} \) becomes the standard Anderson form,

\[ \tilde{H} = \sum_\sigma \varepsilon_\sigma \hat{d}_\sigma \hat{d}_\sigma^\dagger + U \hat{d}_{\sigma L} \hat{d}_{\sigma L}^\dagger \hat{d}_{\sigma R} \hat{d}_{\sigma R}^\dagger + \sum_{k,\sigma,\alpha} \varepsilon_k \hat{C}_{k\sigma\alpha}^\dagger \hat{C}_{k\sigma\alpha} + \sum_{k,\sigma,\alpha} (V_{\alpha} \hat{C}_{k\sigma\alpha}^\dagger \hat{d}_\sigma + \text{H.c.}), \]

but with the negative bipolaronic interaction \( U \). Below we address the transport properties of QDs described by this Hamiltonian.

Within the Keldysh formalism [15], the electronic current through our system in the wide-band limit is \( I = \frac{e}{2} \text{Im} \langle f_{R}(\varepsilon) - f_{L}(\varepsilon) \rangle \sum_{\sigma} \text{Im} G_{\sigma}^{\text{R}}(\varepsilon) \), which expresses the current as an integral of the elastic transmission probability over the conduction band weighted by the difference of the Fermi functions \( f_{\alpha}(\varepsilon) \) in the two leads. The transmission probability is constructed in terms of the dot retarded Green function \( G_{\sigma}^{\text{R}}(\varepsilon) \) and the dot level broadening due to tunnel coupling to the leads, \( \Gamma = \sum_{\alpha} \Gamma_{\alpha} = \pi \sum_{k} |V_{\alpha}|^2 \delta(\mu_{\alpha} - \varepsilon_{k}) \) calculated at the lead Fermi energy \( \mu_{\alpha} \).

To determine the Green function \( G_{\sigma}^{\text{R}}(\varepsilon) \), we employ the equation-of-motion (EOM) approach [5,14–19]. The EOM for \( G_{\sigma}^{\text{R}}(\varepsilon) \) gives rise to higher-order Green functions, whose EOMs generate, in turn, more higher-order ones. Provided that proper decoupling procedures have been used to truncate this hierarchy, the approach can work in all parameter regimes. For our purpose, we neglect those higher-order Green functions which involve the spin-exchange scattering and simultaneous creation or annihilation of two electrons in the QD orbital, thereby excluding the contributions from the Kondo effect, pair tunneling and cotunneling [14,19]. By this scheme, we derive the dot Green function as

\[ G_{\sigma}^{\text{R}}(\varepsilon) = \frac{1 - n_{\sigma}}{\varepsilon - \varepsilon_{\sigma} + i\Gamma} + \frac{n_{\sigma}}{\varepsilon - \varepsilon_{\sigma} - U + i\Gamma}, \]

where the occupation number \( n_{\sigma} = \langle \hat{d}_{\sigma}^\dagger \hat{d}_{\sigma} \rangle \) should be calculated from the nonequilibrium lesser Green function for the dot, \( n_{\sigma} = -\langle \hat{b}^\dagger \hat{b} \rangle \). Applying the formal Keldysh Green-function technique [15] to our system, the lesser Green function is related to the retarded one through \( G_{\sigma}^{\text{R}}(\varepsilon) = -2i/\Gamma \sum_{\alpha} \Gamma_{\alpha} f_{\alpha}(\varepsilon) \text{Im} G_{\sigma}^{\text{R}}(\varepsilon) \). We can thus self-consistently calculate \( n_{\sigma} \) and \( G_{\sigma}^{\text{R}}(\varepsilon) \), from which

\[ \text{Taking the average value on the phonon state means that dynamics of the phonon cloud is averaged accounted for and fluctuations of the boson fields are neglected, which is valid for strong electron-phonon interaction } M \gg V_{\alpha} \text{ favoring the formation of localized polarons [13]. This approximation already gives a good explanation for the phonon sidebands and Franck-Condon blockade effect in the large-bias and high-temperature regime that allows phonon excitations, see, e. g., ref. [14] and references therein.} \]
the current \( I \) or differential conductance \( dI/dV \) can be obtained. Equation (3) shows that in the SE resonant tunneling regime, the QD has two resonances with width \( \Gamma \): one at \( \varepsilon_d \) weighted by \( 1 - n_\sigma \), and one at \( \varepsilon_d + U \) weighted by \( n_\sigma \). A similar solution for \( G^r_{\sigma}(\varepsilon) \) at positive \( U \) was previously derived for explaining the periodic conductance oscillations [5] and charging effects [16] in QDs. Here, we employ eq. (3) to demonstrate the bipolaronic blockade effect due to the bipolaronic attraction between electrons, as a counterpart to the Coulomb blockade physics from the Coulomb repulsion. Despite its simplicity, the involved transport phenomena are unexpected and even surprising.

**Results and discussions.** In the numerical results presented below, the half-bandwidth \( D = 10 \) is taken as the energy unit. We apply a symmetric source-drain voltage \( V \) on the two leads whose Fermi energies are \( \mu_\sigma = \mu + (\delta_\alpha L - \delta_\alpha R) V / 2 \) with \( \mu = 0 \) the equilibrium Fermi energy. The dot level \( \varepsilon_d \) is written as \( \varepsilon_d = \varepsilon_d + (\delta_\alpha L - \delta_\alpha R) B \) in which \( \varepsilon_d \) is the part tuned by the gate voltage and \( B \) is the applied magnetic field. We introduce \( x \equiv \Gamma L / \Gamma R \) to measure the left-right asymmetry of coupling and \( y \equiv 4 x / (1 + x)^2 \) as a dimensionless measure of conductance in the asymmetric case. The temperature \( T = 10^{-8} D \) is set as the smallest energy scale in order to exclude the trivial thermal broadening effect, though it should be higher than the underlying Kondo scale.

We first give, in fig. 1, the dot spectral density \( A(\varepsilon) = -\frac{1}{2 \pi} \sum_{\sigma} \text{Im} G^r_{\sigma}(\varepsilon) \) and the occupation \( n = \sum_{\sigma} n_\sigma \) in the equilibrium case for vanishing magnetic field. As the dot level \( \varepsilon_d \) is tuned downward across the Fermi energy, the spectral weight of the Fermi level \( \varepsilon_d < 0 \) dot shifts to the low-energy side of the Fermi energy more rapidly than that of \( U > 0 \) (figs. 1(a)–(c)), thereby leading to a rapid increase of the occupation (fig. 1(d)). Particularly, the occupation governed by the bipolaronic attraction increases linearly with \( \varepsilon_d \) near the particle-hole symmetric point, where the Coulomb repulsion induces the usual plateau. To give a quantitative account for this difference, we calculate the occupation \( n \) to first order in the deviation from the particle-hole symmetric point \( \delta \varepsilon_d = \varepsilon_d + U / 2 \), which is, in the limit \( T / \Gamma \rightarrow 0 \), \( n = 1 - 8 \Gamma (U^2 + 4 \Delta^2)^{-1} [\pi + 2 \arctan(\pi \delta \varepsilon_d)]^{-1} \delta \varepsilon_d \). For \( \Gamma \ll |U| \), the attraction results in \( n = 1 - \frac{1}{2} \frac{1}{U} \delta \varepsilon_d \) and the repulsion gives \( n = 1 \). Figure 1(d) also indicates that the bipolaronic attraction significantly suppresses the dot spectral density at the Fermi energy as compared with the Coulomb repulsion. These unusual local properties of the \( U < 0 \) dot can bring about very striking transport behavior when the source-drain voltage is applied.

Figure 2(a) presents the conductance map subject to the bipolaronic attraction for symmetric couplings and zero magnetic field, where the conductance lines appear at the thresholds \( V \) and \( \varepsilon_d \) that switch the system between the configurations of two (L-2), one (L-1), and zero (L-0) QD level(s) in the transport window. A striking feature distinct from the usual Coulomb-blockade spectroscopy [1–5] is that the conductance lines under low biases \( |V| < |U| \) is now suppressed. This bipolaronic blockade effect becomes more prominent for weak dot-leading coupling \( \Gamma \) (figs. 2(b)–(f)). Particularly, in the limit \( \Gamma \ll |U| \) the linear conductance is completely suppressed (fig. 2(b)), eliminating the usual periodic Coulomb oscillations at \( \varepsilon_d = 0 \).
π in the middle is estimated to be (strengthening the dot-lead coupling (fig. 3(c)). Its height increasing the asymmetry (fig. 3(b)) and is elevated by into finite plateaus. The plateau becomes concrete for $x<1$ regions with positive (or negative if $x>1$) slope merge (fig. 3(a)) for the transitions between the L-1 and L-2 configurations, $U_{L-1}$ configuration where the negative transport window, the tunneling is also suppressed in the $L-0$ configuration since there has no QD levels in the configurations. While SE tunneling is obviously not allowed in small spectral weight left in the transport window. There- (fig. 2(g)) or doubly occupied (fig. 2(h)) QD with very $\epsilon_{d}$ and $|U|$. Note that the conductance lines at $|V| < |U|$ represent the transitions between the L-0 and L-1 configurations. While SE tunneling is obviously not allowed in the L-0 configuration since there has no QD levels in the transport window, the tunneling is also suppressed in the L-1 configuration where the negative $U$ favors an empty (fig. 2(g)) or doubly occupied (fig. 2(h)) QD with very small spectral weight left in the transport window. Therefore, no significant current change occurs between the two configurations, leading to the suppressed conductance at low biases. On the other hand, the L-2 configuration always allows two independent SE tunnelings (fig. 2(i)) giving the maximal current $I_{0} \equiv g\epsilon / h$. As a result, the conductance lines for large biases $|V| > |U|$, representing the transitions between the L-1 and L-2 configurations, are thus enhanced.

When asymmetric coupling is introduced, as shown in fig. 3(a) for $x > 1$, the conductance lines enclosing the L-1 regions with positive (or negative if $x < 1$) slope merge into finite plateaus. The plateau becomes concrete for increasing the asymmetry (fig. 3(b)) and is elevated by strengthening the dot-lead coupling (fig. 3(c)). Its height in the middle is estimated to be $(\pi \Gamma / |U|)g\epsilon^{2} / h$ in the asymmetric limit. To explain the formation of the plateau, let us consider a given electronic state $\epsilon_{d} = |U|$. As the

![Fig. 3](image)

Fig. 3: (Colour on-line) (a) Conductance map for asymmetric coupling $\Gamma = 0.1|U|$, $x = 20$. (b) and (c): conductance vs. $V$ for different $x$ with $\Gamma = 0.1|U|$ and different $\Gamma$ with $x = 500$, respectively. (d) Occupation number vs. $V$ for different $x$ with $\Gamma = 0.1|U|$. (e) and (f): schematic diagrams for the shift of QD spectral weight in the asymmetric limit when $V$ increases positively (e) and negatively (f), where the blue (orange) lines corresponds to the spectral densities at equilibrium (at indicated $V$). Parameters: $\epsilon_{d} = |U|$ for (b)–(f), $U = -0.1$, $B = 0$. Source-drain bias rise positively, the occupation number $n$ increases linearly for $x \gg 1$ (fig. 3(d)), since in this limit $n$ is only determined by the left lead and is equivalent to the equilibrium one (fig. 1(d)). Accordingly, the QD spectral weight shifts linearly from the level $\epsilon_{d}$ to the level $\epsilon_{d} + U$ which lies in the transport windows (fig. 3(e)) giving rise to a linear increase of the current. Therefore, the conductance is constant until the Fermi level of the left lead $\mu_{L}$ sweeps over the level $\epsilon_{d}$ saturating the current. On the other hand, as the bias rises negatively, the QD spectral density in the transport window and hence the current keep to be nearly zero until $\mu_{R}$ sweeps over the level $\epsilon_{d}$ (fig. 3(f)), which leads to the conductance peak at $V = -2|U|$ (see figs. 3(a)–(c)).

![Fig. 4](image)

Fig. 4: (Colour on-line) Conductance map under magnetic field $B = 0.15|U|$ for (a) symmetric coupling $x = 1$ and (b) asymmetric coupling $x = 20$. (c) Lines cut from (b) at $\epsilon_{d} = |U|$ for different magnetic fields and $x = 500$. (d) Amplitude ratio of magnetic-split conductance peaks as a function of the coupling asymmetry. In (a) and (b), the squares, circles, and triangles denote three representative biases at $\epsilon_{d} = |U|$. Other parameters: $\Gamma = 0.1|U|$ for (a)–(c) and $U = -0.1$.

We now turn to examine the effect of magnetic field, which splits the conductance peaks and narrows the plateaus (figs. 4(a)–(c)). While the narrowing of the plateaus is quite straightforward by just noting that at fixed $\epsilon_{d}$ and $x \gg 1$ the plateau starts and ends when $\mu_{L}$ crosses the levels $\epsilon_{\uparrow} + U$ and $\epsilon_{\downarrow}$, the feature of split conductance peaks deserves a careful analysis. For symmetric couplings, the amplitudes of split peaks are different, reaching a ratio of 3 in the weak-coupling limit (fig. 4(d)). This is different from the Coulomb blockade physics where the corresponding ratio is 2 [20]. The underlying mechanism can be revealed by analyzing the energy-level diagrams of $5$ at $\epsilon_{d} = |U|$ and characteristic biases marked by squares, circles, and triangles in figs. 4(a) and (b). Note that at finite magnetic field the spectral weights of the four QD levels $\epsilon_{\uparrow}$, $\epsilon_{\downarrow}$, $\epsilon_{\uparrow} + U$, and $\epsilon_{\downarrow} + U$ are proportional to

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I = 0 fore, two conductance peaks with the amplitude ratio of the occupation $n_1 = 1$ ((a)–(c)) and asymmetric $x > 1$ ((d)–(f)) couplings with finite magnetic field, under the source-drain biases corresponding to the squares ((a) and (d)), circles ((b) and (e)), and triangles ((c) and (f)) in fig. 4. The light-gray, gray, and dark-gray lines represent the four QD levels with small, moderate, and large spectral weights, respectively.

The most favorable candidate to cause a be to use suspended carbon-nanotube devices, for which negative charging energy. A conceivable realization might on the observability of these transport phenomena for finite magnetic field, under the source-drain biases corre-

sponding to the squares ((a) and (d)), circles ((b) and (e)), and triangles ((c) and (f)) in fig. 4. The light-gray, gray, and dark-gray lines represent the four QD levels with small, moderate, and large spectral weights, respectively.

the occupation $1 - n_4, 1 - n_7, n_1,$ and $n_7$, respectively, as shown by eq. (3). Since $n_σ ≃ 0$ for the negative bias at the square point (fig. 5(a)), no significant spectral density is enclosed in the transport window giving $I ≃ 0$. As the bias further decreases to the circle point (fig. 5(b)), one has $n_7 ≃ 0.25$ and $n_1 ≃ 0.5$. This produces a current $I = 0.75I_0$, in which the portion $0.5I_0$ is the contribution from the two spin-down levels $ε_↓$ and $ε_↓ + U$, and $0.25I_0$ from the spin-up level $ε_↑ + U$. The current finally saturates at $I_0$ under the bias at the triangle point where the total QD spectral density contributes (fig. 5(c)). Therefore, two conductance peaks with the amplitude ratio of 3 appear as $μ_ε$ successively sweeps over the Zeeman-split levels $ε_↓$ and $ε_↑$. Similar analysis can indicate that the ratio turns to 1 in the asymmetric limit (fig. 4(d)). In this case, the occupation is only determined by the stronger coupled lead and one always have $n_σ ≃ 0$, which results in the current $I ≃ 0$ (fig. 5(d)), $0.5I_0$ (fig. 5(e)), and $I_0$ (fig. 5(f)) at the three characteristic biases, respectively.

Final remarks. – Finally, it is helpful to comment on the observability of these transport phenomena for negative charging energy. A conceivable realization might be to use suspended carbon-nanotube devices, for which strong electron-phonon coupling has already been observed [7]. The most favorable candidate to cause a negative $U$ in nanotubes is the longitudinal stretching mode with the maximal dimensionless coupling $λ ≃ \frac{U}{ε_p} ≃ 0.01 \text{meV} \cdot \text{nm}^{-1}$ [21] and the phonon energy $ε_p ≃ 0.1 \text{meV}$ [22], where $L_⊥$ is the nanotube circumference and $L$ the length. Using $L_⊥ \sim 1 \text{nm}$ and $L \sim 100 \text{nm}$, we estimate the polaronic energy shift as $2L^2ε_p \sim 20 \text{meV}$, being of the same order as or larger than typical Coulomb repulsion in nanotube QDs [23]. After realizing $U < 0$, transport spectroscopy measurements similar to the usual Coulomb blockade technique are enough to verify our predictions, including the suppression of conductance at low biases, the appearance of conductance plateaus for asymmetric coupling, and the ratio of 3 for magnetic-split conductance peaks.

Although we have made these predictions for the low biases and temperature that prohibit vibrational excitations, the effect of phonon sidebands on our results, due to phonon excitations in the large-bias and high-temperature regime, can be easily expected. For example, since the conductance plateau and phonon sidebands appear respectively at bias voltages of the order of $|U|$ (see fig. 3) and larger than $ε_p$ (see, e.g., ref. [14]), they are well separated in the conductance map for $ε_p ≫ |U|$ and thus the plateau is robust against the phonon excitation. On the contrary, if $ε_p \sim |U|$ or $ε_p \gg |U|$, the sidebands appear in the same region where the plateau shows up, thereby distorting the plateau. For large temperature, the plateau is also subject to a thermal broadening effect. As shown by fig. 3, while the plateau width is determined by $|U|$, the width of the plateau border is determined by $Γ$. Therefore, if $T > Γ$, the border of the plateau will be smeared out. For $T > |U|$, even the entire plateau could be smeared out.

We hope that these remarks could help as a guide to tailoring relevant experimental parameters for observing our predictions.

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