Coherent Three-Level Mixing in an Electronic Quantum Dot

C. Payette,[1,2] G. Yu,[1] J. A. Gupta,[1] D. G. Austing,[1,2,*] S. V. Nair,[3] B. Partoens,[4] S. Amaha,[5] and S. Tarucha[5,6]

[1]*Institute for Microstructural Sciences M50, NRC, Montreal Road, Ottawa, Ontario, K1A 0R6, Canada*
[2]*Department of Physics, McGill University, 3600 rue University, Montréal, Quebec, H3A 2T8, Canada*
[3]*Center for Advanced Nanotechnology, University of Toronto, 170 College Street, Toronto, Ontario, M5S 3E3, Canada*
[4]*Departement Fysica, Universiteit Antwerpen, Groenenborgerlaan 171, B-2020 Antwerpen, Belgium*
[5]*Quantum Spin Information Project, ICORP, JST, Atsugi-shi, Kanagawa 243-0198, Japan*
[6]*Department of Applied Physics, University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*
(Received 15 August 2008; published 16 January 2009)

We observe magnetic-field-induced level mixing and quantum superposition phenomena between three approaching single-particle states in a quantum dot probed via the ground state of an adjacent quantum dot by single-electron resonant tunneling. The mixing is attributed to anisotropy and anharmonicity in realistic dot confining potentials. The pronounced anticrossing and transfer of strengths (both enhancement and suppression) between resonances can be understood with a simple coherent level mixing model. Superposition can lead to the formation of a dark state by complete cancellation of an otherwise strong resonance, an effect resembling coherent population trapping in a three-level-system of quantum and atom optics.

DOI: 10.1103/PhysRevLett.102.026808 PACS numbers: 73.21.La, 73.22.Dj, 73.23.Hk

Energy level mixing leading to level anticrossing, and superposition of states, are key quantum properties in electronics and optics in the solid state. Mixing of two levels is prevalent in semiconductor nanosystems [1–4]. Mixing of two levels or excitations of different origin or character commonly leads to monotonic exchange of resonance character between two anticrossing branches as a function of an external parameter. An example is electric-field induced mixing of direct (bright) and indirect (dark) exciton states in coupled quantum dots [1].

Here we demonstrate a means to realize dark state formation by three-level mixing. This is potentially a highly useful tool for quantum coherent transport involving multiple quantum levels [5–7]. Few-electron properties of single vertical dots in gated devices reveal that the lateral confinement is nearly elliptical [ellipticity (δ) $\sim$ 1.05 to 2] and parabolic [8–10]. For single electrons resonantly traversing a quantum dot molecule we take advantage of this high but not perfect symmetry in the dot confinement potential to electronically mix single-particle levels brought into proximity with a magnetic (B) field. Avoided two level crossings in excitation spectra of single and coupled dots have been observed by transport in the few- and many-electron ($N > 1$) regimes, where Coulomb interactions are a nontrivial factor [3,4], but mixing of three (or more) levels is required for advanced quantum information protocols [5–7].

With two weakly coupled vertical dots we can access many single-particle states of the individual dots over a large energy window. From a $GaAs/Al_{0.22}Ga_{0.78}As/In_{0.05}Ga_{0.95}As$ triple-barrier structure we fabricate a gated submicron circular mesa [11]. The two dots, strongly confined in the vertical direction, are weakly coupled to the doped contacts through 8.5 nm outer barriers, and weakly coupled to each other through an 8.5 nm central barrier (tunnel coupling, Δ_{SAS}, <0.1 meV, is sufficiently weak that it plays no important role in our main results). Current flows through the dots in response to a bias, V_{sd}, between the top contact and substrate, and a voltage on the gate, V_g. With a B field applied parallel to the current, we can manipulate the energy position of the single-particle states in both dots. We use the 1s-like ground state in the emitter (upstream) dot to probe single-particle states of the collector (downstream) dot in the single-electron elastic resonant tunneling regime (Fig. 1) [12].

For an ideal two-dimensional (2D) effective lateral confinement potential that is strictly elliptical and parabolic [$V_{\rm eff}(x, y)\alpha(\delta x^2 + \delta^{-1}y^2)$ for $\delta \geq 1$, see Ref. [13]] the single-particle states of the dot not only evolve with B field in a distinct way, but all level crossings are exact crossings as exemplified by spectra in Figs. 1 and 2(b). Counter to this, we commonly observe pronounced anticrossing behavior between two or more approaching energy levels in probed dot spectra even though overall the spectra are well accounted for by calculations assuming ideal confinement. Focusing discussion on one entirely typical device, we argue this is due to mixing brought about by deviations from ideal lateral confining potentials in realistic dots [14].

The measured high energy spectrum in Fig. 2(a) and selected current traces in Fig. 2(c) demonstrate that three-level mixing is readily observed [14]. The spectrum is well fitted overall by the calculated spectrum in Fig. 2(b) for a 2D elliptical parabolic dot ($\delta \sim 4/3$), even though the dot mesa is circular [15,16]. The three lowest energy and very

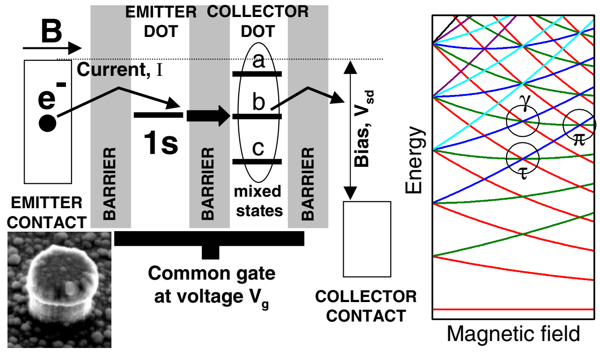

FIG. 1 (color online). Measurement principle: energy separation between the $1s$ state in the emitter dot and mixed states (a, b, c) in the collector dot is a function of (V_{sd}, V_g) ($1s$-b resonance condition shown). Empty nonresonant states below (a, b, c) omitted. Micrograph is of mesa similar to measured mesa. Lowest three exact three-level-crossings are circled in the Fock-Darwin spectrum for circular parabolic dot. Energy of states is with respect to energy of the $1s$ state. States that merge into the same Landau level at high B field are colored the same.

distinctive three-level crossings, which we label τ, γ, and π (Fig. 1), occur at finite B field and are robust features although $\delta \neq 1$.

What is the origin of the level mixing not reproduced by the single-particle calculation? The observed level splitting energy is typically 100's μ eV ($\gg k_b T \sim 25$ μ eV) and as large as $\sim$1 meV (our spectral resolution is $\sim$50 μ eV). It is much larger than other sources of level splitting: Δ_{SAS}, Zeeman splitting (<100 μ eV up to 4 T) [17], and spin-orbit splitting (<50 μ eV) [17]. Since the spectrum is single particle in nature, splitting due to Coulomb interactions can be discounted [4]. We therefore attribute the behavior near the crossings to non-negligible higher-degree terms (nth degree terms $x^p y^q$ with $n = p + q > 2$) in the probed dot's confining potential $V_{\rm eff}(x, y)$ due to anharmonicity and anisotropy arising from natural randomness and local imperfections [12]. Confining potentials of real dots always vary to a degree and nonsystematically from device to device and even dot to dot in multidot devices.

Although specific details of the source of the mixing remain unclear, we still exploit it. A simple 3×3 matrix Hamiltonian model allows us to examine the region where three levels cross in the presence of a symmetry breaking potential. Near the common crossing point three initially uncoupled basis levels, numbered 1–3 in Fig. 2(d), are assumed to have a linear dispersion with B field, a good approximation in the vicinity of a crossing. The couplings between each pair of these levels are characterized by three off-diagonal matrix elements (coupling energies), C_{12}, C_{13}, and C_{23}. They are assumed to be real, but may be positive or negative, and independent of B field and of each other. Cartoons of the four basic types of three-level crossings predicted by the model are given in Fig. 2(d) [18]. Candidate exact crossings are rarely observed. Instead,

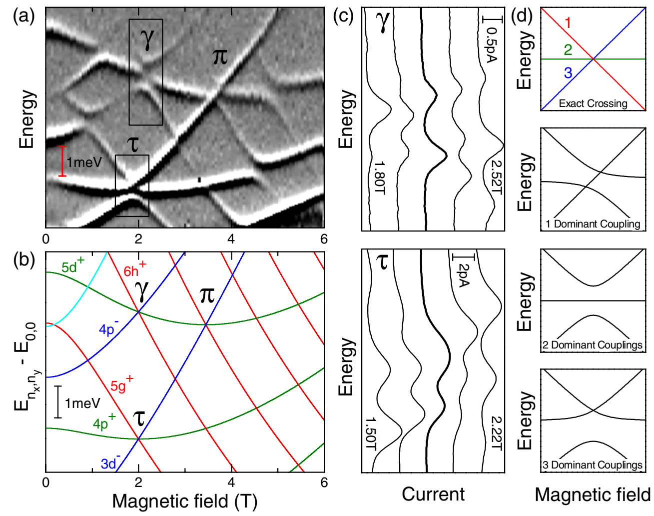

FIG. 2 (color online). (a) Differential conductance resonance position versus B field maps out the high energy spectrum of the probed dot at 0.3 K [15,16]. (b) Calculated spectrum with confinement strengths $\hbar\omega_x = 6.1$ meV and $\hbar\omega_y = 4.6$ meV ($\delta = \omega_x/\omega_y \sim 4/3$) reproduces a well the measured spectrum except in the vicinity of level crossings. Energy of the (n_x, n_y) state is with respect to the energy of the (0,0) ground state (n_x and n_y are x- and y- quantum numbers [13]). Relevant states are labeled with atomic orbital-like notation for circular dot. (c) Selected current traces (vertical sections) through the γ and τ crossings from the boxed regions in panel (a) [nonresonant component not subtracted]. (d) Basic types of three-level crossings. Dimensionless couplings are set to $C_{12} = C_{23} = C_{13} = 0$; $C_{12} = 1$, $C_{23} = C_{13} = 0$; $C_{12} = C_{23} = 1$, $C_{13} = 0$; and $C_{12} = -1$, $C_{23} = C_{13} = 1$.

more interesting one-, two-, and three-dominant coupling-type crossings are seen. In Fig. 2(a), crossings π, γ, and τ appear to be, respectively, one-, two-, and three-dominant coupling-type crossings. With our model we can also calculate the currents through the coupled states of the downstream dot.

We focus now on three-level crossing γ as it clearly illustrates mixing leading to bright resonance-to-dark resonance interconversion [18]. The measured B field dependence of the energy levels is given in Fig. 3(a) in the vicinity of the crossing between the $4p^-$-, $5d^+$-, and $6h^+$-like basis states [$(n_x, n_y) = (2, 1)$, (1,3), and (0,5) respectively]. Its distinctive shape is due to two dominant and approximately equal couplings between the $6h^+$- and $5d^+$-like states (C_{12}), and between the $5d^+$- and $4p^-$-like states (C_{23}), whereas the coupling between the $6h^+$ and $4p^-$-like states (C_{13}) is very weak. Mixing leads to the conversion of two dark (upper and lower) resonances and one bright (center) resonance to the left of the crossing to one dark resonance and two bright resonances at the crossing [Fig. 3(b)]. The center resonance current is zero at $\sim$2.2 T when the branches are minimally separated. This cancellation is a strong signature of interference. To the right of the crossing the center resonance recovers its

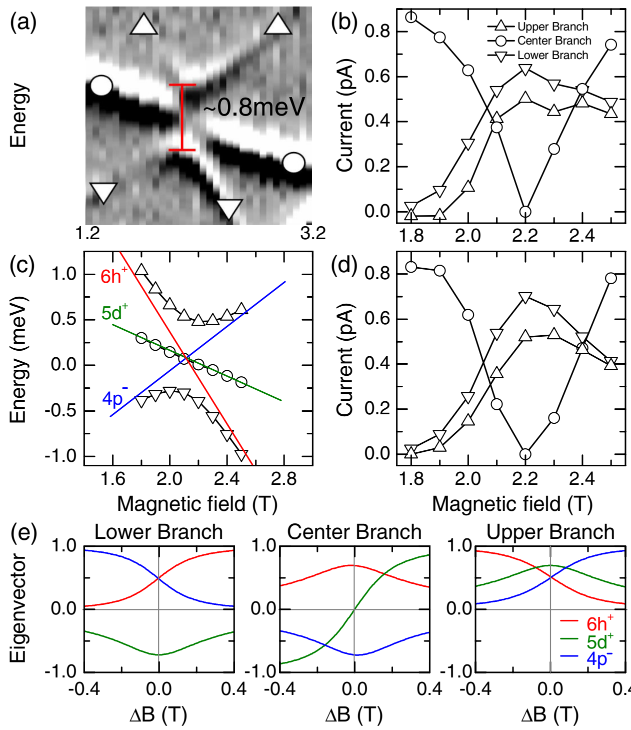

FIG. 3 (color online). Three-level-crossing γ. (a) Energy level (differential conductance resonance) positions. (b) Extracted branch currents (resonance current with nonresonant component subtracted). (c) Fit of energy level positions: couplings (in meV) $C_{12} = 0.30$, $C_{23} = 0.29$, and $C_{13} = 0.03$. Lines estimating position of uncoupled basis levels provide a guide to the eye. (d) Fit of currents. Resonances through uncoupled basis states have amplitudes (in $pA^{1/2}$) $s_1 = -0.4$, $s_2 = 0.804$, and $s_3 = 0.219$ at 1.8 T, and $s_1 = -0.213$, $s_2 = 1.22$, and $s_3 = 0.209$ at 2.5 T. (e) Reconstructed eigenvectors. $\Delta B = 0$ T is at ~2.12 T.

strength and the upper and lower resonances start to weaken.

The resonant currents through the basis states may be described by current amplitude parameters (s) that represent the tunnelling amplitudes through the uncoupled states. We denote these parameters by s_1, s_2, and s_3, one for each of the uncoupled states. When squared these give approximately the branch currents far to the left and right of the crossing where the states are essentially uncoupled. In a simple coherent tunneling picture derived from a Fermi golden rule argument applicable for weak tunnel coupling, we compute each branch current by squaring the sum of the eigenvector (v) components multiplied by the current amplitudes (s). Explicitly, $I_j = c|\langle g|\psi_j\rangle|^2 = |\sum_m v_m^j s_m|^2$, where I_j is the resonance current of the j = (upper, center, lower) branch, $m = 1, 2, 3$ is the basis state index, c is a constant, g is the 1s-like ground state of the upstream dot, and ψ_j is the branch state of the downstream dot. $\langle g|\psi_j\rangle$ is an in-plane overlap integral. A simultaneous least squares fit of the measured energy level position and resonance current is given in Figs. 3(c) and 3(d). In the simplest model one may take the current amplitudes (s) to be independent of B field. However, this does not reproduce the data well. We found that a simple linear variation of s with B field adequately accounts for the intrinsic variation of the resonance current through each of the three states in the vicinity of the crossing. This is true for all crossings studied. Thus we use six values of s, three each representing current amplitudes at ~0.35 T to the left and right of the crossing. The uncoupled current amplitudes at any B field are then obtained by a linear interpolation between these left and right values.

To understand how mixing gives rise to the observed branch currents, we reconstruct in Fig. 3(e) the eigenvectors for the three branches giving the components of the uncoupled basis states. Well to the left and right of the crossing the expected behavior is apparent. For each branch one of the three components tends to ± 1 and the remaining two tend to zero meaning the branch current simply tends to the square of the relevant current amplitude. At the crossing point of the uncoupled basis levels ($\Delta B = 0$ T), for $C_{12} \sim C_{23} \gg C_{13}$, the currents are approximately given by the following expressions: $I_{\mathrm{upper}} \sim \frac{1}{4}(s_1 + \sqrt{2}s_2 + s_3)^2$, $I_{\mathrm{center}} \sim \frac{1}{2}(s_1 - s_3)^2$, and $I_{\mathrm{lower}} \sim \frac{1}{4} \times (s_1 - \sqrt{2}s_2 + s_3)^2$. Since $|s_2| > |s_1| \sim |s_3|$, I_{upper}, $I_{\mathrm{lower}} \sim \frac{1}{2}(s_2)^2$ and I_{center} is greatly reduced from its uncoupled value of $\sim(s_2)^2$. Thus, the upper and lower branch resonances are dominated by the large contribution of the uncoupled center resonance current amplitude, s_2, and appear bright. Furthermore, the center branch resonance appears dark when the influence of s_2 is zero. I_{center} will vanish precisely at $\Delta B = 0$ T if s_1 and s_3 are equal in magnitude and of the same sign. The position of the current cancellation can be shifted slightly from $\Delta B = 0$ T if certain values of C and s are not the same. For the γ crossing, s_1 and s_3 while of similar magnitude are of opposite sign at $\Delta B = 0$ T so a small contribution of s_2 is required to attain zero center branch current. This occurs a little to the right of $\Delta B = 0$ T. The current suppression and cancellation are thus genuine and robust effects arising from three-level mixing.

The quantum-electronic effect in Fig. 3 bears strong similarity with coherent population trapping in three-level systems of quantum and atom optics [19]. When two states are optically driven to a common excited state a nonabsorption resonance occurs due to destructive interference between two absorption pathways [20], and a dark state results identical in form to the center branch eigenvector at $\Delta B = 0$ T in Fig. 3(e). Two all-electrical single-electron tunneling schemes, one for coherent population trapping and one for coherent tunneling by adiabatic passage, have been proposed for three triangularly or linearly arranged dots [5,6]. Our situation of three levels mixing intradot is equivalent to three levels, one per dot, mixing interdot envisaged by these schemes (also see [7]).

Finally, we present a numerical study of a symmetry breaking potential to explain the mixing at the γ crossing. Figure 4 shows calculations replicating well the observed

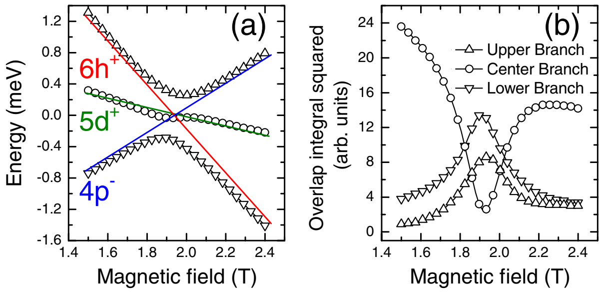

FIG. 4 (color online). Calculations reproducing correct general behavior at γ crossing in Fig. 3. (a) Energy level position, and (b) overlap integral squared, proportional to resonance current in coherent tunneling picture, $|\langle g|\psi_j\rangle|^2$, for each branch. Parameters: Probed dot's $V_{\rm eff}(x, y) = 1/2[(4/3)x^2 + y^2 + 0.12x^2y + 0.05xy^2 + 0.05y^3 + 0.01x^6 + 0.01y^6]$, $\delta = 4/3$ and $\hbar\omega_y = 3.8$ meV. Emitter dot potential is the same but 3rd and 6th degree terms are excluded (1s-like prober state is least sensitive to addition of perturbation terms).

behavior. Some higher-degree terms in $V_{\rm eff}(x, y)$ of strength up to $\sim$15% are included, specifically dominant 3rd degree terms, x^2y, xy^2, and y^3, and weaker 6th degree terms, x^6 and y^6. The former (latter) effectively account for the two dominant couplings, $C_{12} \sim C_{23}$ (much weaker coupling, C_{13}) [21]. The center branch resonance is strongly suppressed when the probed mixed state (a linear combination of basis states) in the downstream dot is nearly orthogonal to the 1s-like prober state in the upstream dot.

We thank A. Bezinger, D. Roth, and M. Malloy for assistance, and K. Ono, T. Kodera, T. Hatano, Y. Tokura, M. Stopa, M. Hilke, G. C. Aers, M. Korkusinski, and R. M. Abolfath for discussions. Part of this work is supported by NSERC (Discovery grant 208201), Flemish Science Foundation (FWO-VI), Grant-in-Aid for Scientific Research S (No. 19104007), B (No. 18340081), SORST-JST, Special Coordination Funds for Promoting Science and Technology, and MEXT.

*guy.austing@nrc-cnrc.gc.ca

[1] H. J. Krenner *et al.*, Phys. Rev. Lett. **94**, 057402 (2005); E. A. Stinaff *et al.*, Science **311**, 636 (2006).
[2] L. Zimin, S. V. Nair, and Y. Masumoto, Phys. Rev. Lett. **80**, 3105 (1998); B. A. Carpenter *et al.*, Phys. Rev. B **74**, 161302(R) (2006); S. F. Fischer *et al.*, Nature Phys. **2**, 91 (2006); K. Hennessy *et al.*, Nature (London) **445**, 896 (2007).
[3] T. H. Oosterkamp *et al.*, Phys. Rev. Lett. **80**, 4951 (1998).
[4] C. Ellenberger *et al.*, Phys. Rev. Lett. **96**, 126806 (2006).
[5] B. Michaelis, C. Emary, and C. W. J. Beenakker, Europhys. Lett. **73**, 677 (2006).
[6] A. D. Greentree *et al.*, Phys. Rev. B **70**, 235317 (2004).
[7] M. Korkusinski *et al.*, Phys. Rev. B **75**, 115301 (2007).
[8] S. Tarucha *et al.*, Phys. Rev. Lett. **77**, 3613 (1996).
[9] P. Matagne *et al.*, Phys. Rev. B **65**, 085325 (2002).
[10] T. Fujisawa *et al.*, Nature (London) **419**, 278 (2002).
[11] D. G. Austing *et al.*, Physica (Amsterdam) **249–251B**, 206 (1998).
[12] K. Ono *et al.*, Physica (Amsterdam) **314B**, 450 (2002); C. Payette *et al.*, Physica (Amsterdam) **40E**, 1807 (2008); S. Amaha *et al.*, Phys. Status Solidi C **5**, 174 (2008).
[13] This is a common assumption in the modeling of dots when the vertical (z-) confinement is much stronger. See, e.g., S. M. Reimann and M. Manninen, Rev. Mod. Phys. **74**, 1283 (2002); A. V. Madhav and T. Chakraborty, Phys. Rev. B **49**, 8163 (1994).
[14] We studied seven dot spectra in detail from four devices and all exhibited widespread anticrossing behavior at level crossings. Pronounced (well resolved) anticrossing was seen at 11 out of 14 three-level crossings examined.
[15] The energy axis in Fig. 2(a) corresponds to a covariation of V_{sd} and V_g (full details in Ref. [12]). Over the $\sim$10 meV window shown the relation between energy and voltage is almost linear. The energy scale is estimated by looking at the B field position of the level crossings and the relative level spacings and fitting to a standard elliptical parabolic spectrum to extract the confinement strengths.
[16] The absence of extra spectral features in Fig. 2(a), the close overall agreement between the measured spectrum and that calculated in Fig. 2(b)[energies plotted with respect to that of (0,0) ground state], and B field dependence of 1s-1s resonance (not shown) confirm that only the 1s emitter state is involved.
[17] J. Könemann *et al.*, Phys. Rev. Lett. **94**, 226404 (2005).
[18] In Fig. 2(d), basis level 2 has zero gradient while basis levels 1 and 3 have gradients of equal magnitude but opposite sign, and not all possible variants of basic shapes are shown. Our full model allows for arbitrary gradient of basis levels, basis levels not meeting exactly at one point, and free choice of C number. Treating values of C and s as real (positive or negative) numbers appeared adequate for nearly all crossings studied. The model successfully explains mixing at several other two- and three-level crossings. Strong branch current suppression observed at other three-level crossings (not shown) occurs for conditions other than those relevant to the case study of Fig. 3; i.e., dark states do not arise from unique circumstances.
[19] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. **70**, 1003 (1998); M. O. Scully, and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, England, 2006).
[20] C. Santori *et al.*, Phys. Rev. Lett. **97**, 247401 (2006).
[21] The lowest degree monomial term in $V_{\rm eff}$ that can induce direct mixing between any two single-particle elliptical-parabolic-dot states with quantum numbers (n_{x1}, n_{y1}) and (n_{x2}, n_{y2}) is of degree $m = |n_{x1} - n_{x2}| + |n_{y1} - n_{y2}|$. For $6h^+$ $(n_x, n_y) = (0, 5)$ to mix with $5d^+$ (1,3), and $5d^+$ to mix with $4p^-$ (2,1), 3rd degree terms are required since $m = 3$ in both cases. For $6h^+$ to mix with $4p^-$, 6th degree terms are required since $m = 6$. The higher-degree terms included in the calculations in Fig. 4 are chosen to reproduce the general B field dependence of both the energy levels and resonance currents but are restricted to only those terms that could couple the constituent basis states. Third and 6th degree terms other than those included may play a role.