Isotopic disorder in integer and fractional quantum Hall effects

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The energy of the activation gaps in the quantum Hall effect has been thought to be reduced by the broadening of the Landau levels due to disorder. The isotopic mass can affect the electron-phonon interactions and the 0 K renormalization of the conduction and valence bands, leading to appreciable energy offsets between lattice sites with different nuclear masses. The isotopic disorder originating from the natural occurrence of nuclear masses could be the dominant broadening mechanism remaining, leading to the experimentally observed quantum Hall effect gaps in high-mobility devices. The Landau level broadening due to the isotopic disorder has been calculated microscopically without fitting parameters.

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I. INTRODUCTION

Shortly after the discovery of the quantum Hall effect [1,2] and the postulation of the quasiparticle wave function by Laughlin [3], it became clear that disorder plays an important role [4–11]. Disorder broadens the Landau levels [12–35], leading to localized regions and reducing the excitation gaps [36–44]. The disorder-induced broadening is thought to be mainly due to ionized donors separated from the electron gas by a spacer layer of tens of nanometers in width [45]. Additional sources of disorder broadening are chemical impurities in the undoped region and surface roughness that could act as short-range scatterers, and their role was also investigated [44]. Early activation energy calculations and measurements of the odd denominator states provided the first insights into the size of the quasiparticle gaps [46–50]. Rapid improvements in device quality and increased mobility led to the observation of larger excitation energies [36]. However, as the mobilities continued to increase, only modest increments of the excitation gaps were measured.

In order to account for the discrepancies between the observed and predicted excitation gaps, additional factors that could cause a reduction in the gap size were included in modeling. The finite width of the Landau level wave function was taken into account, together with the mixing of the Landau levels [51–53]. While these factors improved the prediction of the gaps, still, discrepancies remained [40]. The discrepancies between measurements [43,44,54–56] and calculations [40,57] are even larger for the smaller excitation gaps of even-denominator states, raising further questions about the role of disorder in obscuring the underlying physics of the $5/2$ state.

The effect of the isotopic disorder, or the natural occurrence of different isotopic masses on different lattice sites in the undoped GaAs region, may appear negligible at first. The lack of differences in charge between isotopic species and the modest changes in lattice constant due to different isotopic masses makes the perturbing effect of the isotopes appear insignificant. However, when the renormalization of the electronic bands due to electron-phonon interactions is taken into account, the size of the effect becomes clear. The 0 K renormalization of the band gap due to electron-phonon interactions can be tens or even hundreds of meV, leading to significant energy offsets between lattice sites occupied by different isotopic species [58–60]. The electron-phonon renormalization is dependent on the nuclear mass, leading to differences in the band gap energy of ∼12.03 K (1.04 meV) per atomic mass unit (amu) in silicon and ∼4.52 K (0.39 meV) per amu in GaAs [58,61].

In this paper, the effect of the isotopic randomness on the Landau levels is calculated. The effect of the minority isotopic species or "isotopic impurities" is considered to be short range and is modeled by a δ-function perturbation. The strength of the perturbation is determined by the energy offset due the 0 K renormalization of the conduction band for the different isotopic masses. The energy offsets between lattice sites break the translational invariance and evade Kohn's theorem, thus leading to a shift of the energy eigenvalue given by the unperturbed Landau level energy E_c for each random distribution of the isotopes within the Landau level wave functions. This energy shift has been calculated for 10^5 random distributions of the isotopic disorder, leading to Gaussian distribution of energies centered around E_c. This study concludes that the isotopic disorder is a remaining appreciable source of Landau level broadening in high-quality quantum Hall devices.

II. ISOTOPIC DISORDER AND ELECTRON-PHONON INTERACTIONS

The change in isotopic composition leads to an energetic shift of the conduction and valence bands of a semiconductor. The energy shift is mainly due to the renormalization of the band energies by the electron-phonon interactions. This many-body effect exists even at 0 K and is responsible for the well-known shift of the electronic band gap with temperature [58,59]. An additional, much smaller contribution to the

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energy shift of the conduction band is due to the shift of the lattice constant with the isotopic composition. The absolute shift of individual bands cannot be measured directly since the usual optical measurements used reveal only the relative shift between the conduction and valence bands.

The microscopic explanation of the temperature-dependent shifts and broadenings of electronic bands in semiconductors can be described by the renormalization of band energies by electron-phonon interactions [62]. A perturbation calculation of the electron self-energy to second order in atomic displacement gives rise two terms, the Fan, or "self-energy," and Debye-Waller terms [63]. The first term corresponds to the first-order electron-phonon interaction using second-order perturbation theory, whereas the second term represents the second-order electron-phonon interaction with first-order perturbation theory. The first term is a self-energy diagram and generates a shift as well as a broadening of the corresponding electronic states, whereas the Debye-Waller term leads only to an energy shift [64].

The electron-phonon interactions depend on the effect of the phonon's vibrating mass. Therefore, the effects arising from the electron-phonon interactions can be divided into two categories: (i) effects that can be described by the average mass, which correspond to the virtual-crystal approximation when more than one isotope of a given atom is present, and (ii) effects in materials with a mixture of several isotopes of a given atom, where mass fluctuations and disorder are important and the virtual crystal approximation breaks down. The virtual crystal approximation introduces an average mass in a crystal with several isotopes in order to recover the translational invariance lifted by the isotopic disorder. However, when the wave function of the electron becomes localized, the random distribution of isotopes becomes important [64].

Effects beyond the virtual-crystal approximation

The effect of the isotopic disorder in solids has been studied over several decades starting with the work of Pomeranchuk, who found that naturally occurring isotopes in crystals will disturb the periodicity of the lattice and thus produce thermal resistance [65]. The isotopic effect on the thermal conductivity was later observed by Geballe and Hull in isotopically enriched germanium [66]. The most commonly used semiconductors are composed of more than one stable isotope in their natural occurrence. In fact, silicon is composed of the three isotopes, ^{28}Si (92.23%), ^{29}Si (4.67%), and ^{30}Si (3.10%), and the most important compound, the semiconductor GaAs, consists of monoisotopic ^{75}As and two Ga isotopes, ^{69}Ga (60.1%) and ^{71}Ga (39.9%). The effects of isotopic composition and disorder on the properties of semiconductors were the subject of numerous more recent studies, as detailed in several reviews [58,59,67].

High-quality crystals of isotopically pure ^{28}Si have revealed surprising effects that are not controlled by the average isotopic composition, but rather by the randomness of the composition present in crystals having natural isotopic abundance. First, it was shown that the isotopic randomness present in natural Si leads to an inhomogeneous broadening that is the dominant factor in determining the observed photoluminescence linewidths of the no-phonon transitions of shallow donor and acceptor bound excitons [68]. Furthermore, the isotopic disorder produces a significant inhomogeneous broadening of many of the long-studied ground to excited state infrared absorption transitions of shallow donors and acceptors [69]. Finally, it was shown that the isotopic randomness present in natural Si is the origin of the much studied ground state splitting of shallow acceptors in Si [70]. The fourfold degenerate acceptor ground state is split by the site-to-site fluctuations of the isotopic mass in the lattice around individual acceptors via the dependence of the valence band edge energy on the isotopic composition, leading to a surprisingly large effect [71,72].

III. RESULTS

The scattering mechanisms that can potentially lower the mobility of a two-dimensional electron gas are well described in the literature [73–75]. Electrons can be scattered by the remote impurities located within the doped region of the device and separated by the spacer layer, as well as residual background impurities in the undoped region. At lower temperature, only phonons with small wave vectors can participate in the process, so that the major contribution to reducing the mobility is expected to come from temperature-independent scattering processes, such as defects and impurities. Alloy-disorder scattering should be negligibly small in high-quality GaAs-based devices, but there is the possibility of scattering by interface charges, located at the heterojunction interface. However, the high perfection and chemical purity that are now achievable in epitaxially grown GaAs, leading to remarkable mobilities at low temperature, raise questions about the possible role of the large isotopic disorder.

The possible effect of isotopic disorder on the electron mobility in the two-dimensional electron gas has not been considered; instead, a different problem will be approached, namely, the perturbing effect of the isotopic disorder on the Landau level states under high magnetic field at very low temperatures. Short-range scattering by chemical impurities in the undoped layer would be quantitatively much stronger than the perturbation induced by the isotopic randomness, but for concentrations low enough, their effect may become negligibly small. The perturbation induced by the screened remote impurities still remains even in the most advanced devices and has been the subject of extensive theoretical studies in the past [11,27,76]. Since this source of perturbation is device specific, a general quantitative comparison with the isotopic perturbation will not be made.

A. Theoretical model

The energy discretization of the conduction band electrons into Landau levels by the magnetic field should not alter significantly the 0 K renormalization by the electron-phonon interactions. Photoluminescence experiments indicate that the Landau levels shift with temperature similar to the way the electronic band gap does [77]. Therefore, the isotopic mass change should have an effect on the Landau levels similar to the one it has on the band gap. However, the energy discretization can lead to intra- and inter-Landau level

scattering, especially when the Landau level separation equals the phonon energies [78–88]. Phonon scattering of the electrons broadens the Landau levels, a process that increases with increasing temperature [32,89].

In order to estimate the effect of the isotopic randomness on the Landau levels, an approach similar to the one that successfully reproduced the isotopic broadening of impurity centers and the ground state splitting of acceptors in Si has been used [71]. The negligibly small strain effects of isotopic substitution have been ignored. Instead, the effect of isotopic composition on the energy of the conduction band has been considered since we are dealing with conduction electrons. The mass dependence of the renormalization of band energies by the electron-phonon interactions via the amplitudes of the zero-point fluctuations varies, on average, as the inverse square root of the average isotopic mass. However, on small length scales, such as the magnetic length l of the Landau levels under high magnetic field [71,72], the statistical distribution of the isotopic species becomes significant, and their effect can be described in terms of fluctuations of the local conduction band energy [68–72]. Such site-to-site energy fluctuations break the translational invariance and evade Kohn's theorem [90–98].

The ^{71}Ga isotopes in natural Ga are treated as isotopic impurities randomly substituted into the perfect ^{69}GaAs. Arsenic has only one stable isotope (^{75}As); therefore, it does not contribute to the isotopic effect. Each ^{71}Ga isotopic impurity is assumed to introduce a short-range δ-function potential. Such perturbing potentials were used previously in order to describe the short-range effect of ionized impurities on the two-dimensional electron gas [12–35]. The local strength of the potential is determined by the energy shift between the conduction bands in ^{69}GaAs and ^{71}GaAs.

The system is described by the Hamiltonian $H = H_0 + V(r)$, where H_0 is the Landau level Hamiltonian,

$$H_0 = \frac{1}{2m_e}[\vec{p} - e\vec{A}(\vec{r})]^2, \tag{1}$$

and $V(r)$ is the disorder potential. Here $\vec{p}$ corresponds to the canonical momentum, and $\vec{A}$ is the vector potential corresponding to the magnetic field $\vec{B}$. In the absence of $V(r)$, the spectrum of H_0 for an infinite system is the known set of equidistant levels, $E_n = (n + 1/2)\hbar\omega_c$, separated by the cyclotron frequency $\omega_c = |e|B/m_e$, where n corresponds to the Landau level index. The Landau states in polar coordinates (r, θ) are given by

$$\varphi_{n,m}(\vec{r}) = C_{n,m}\exp\left((n-m)\theta - \frac{r^2}{4l^2}\right)\left(\frac{r}{l}\right)^{|m-n|} \times L^{|m-n|}_{(n+m-|m-n|)/2}\left(\frac{r^2}{4l^2}\right), \tag{2}$$

with angular momentum m, magnetic length $l = \sqrt{\hbar/|e|B}$, normalization constant $C_{n,m}$, and the Laguerre polynomial $L^n_m(x)$. The perturbing potential induced by the isotopic impurities is given by

$$V(\vec{r}) = \sum_i V_i\delta(\vec{r} - \vec{r}_i), \tag{3}$$

where $\vec{r}_i$ denotes the lattice position of the ith isotopic impurity and V_i is the strength of the perturbation. The isotopic perturbation potential $V(r)$ acts only at the position of the sites of the GaAs crystal that are occupied by ^{71}Ga isotope atoms. Their effect on the Landau level electron at each of these lattice sites is assumed to be proportional to the shift of the bottom of the conduction band between pure ^{71}GaAs relative to ^{69}GaAs. The underlying nature of the perturbing potential is the 0 K renormalization of the conduction band by the electron-phonon interactions, which is modeled as a short-range energy offset perturbing the Landau levels. The value of the energy offset is deduced from experiments. The isotopic potential has been included perturbatively by discretizing the wave function $\phi_n(r)$ on the atomic sites $r = r_i$ and parametrizing the effect of the ^{71}Ga isotopic impurity in $V = \sum_i V_i$ using the simple ansatz

$$\langle\varphi_n(r_i)|V_i|\varphi_n(r_i)\rangle = \delta(r_i)W_i, \tag{4}$$

with

$$W_i = \begin{cases} 0 & \text{for } ^{69}\text{Ga}, \\ \Delta E^{71} & \text{for } ^{71}\text{Ga}. \end{cases} \tag{5}$$

ΔE^{71} represents the energy shift of the minimum of the conduction band in ^{71}Ga As relative to ^{69}GaAs since the electrons occupy a region at the bottom of the conduction band. The energy shift of the conduction band with isotopic mass determines the size of the estimated effect. The quasi-two-dimensional nature of the electron gas is taken into account using the Fang-Howard envelope wave function,

$$\zeta(z) = \left(\frac{b^3}{2}\right)^{1/2} z e^{-bz/2}, \tag{6}$$

where $b = (48\pi m e^2 n^*/\epsilon\hbar^2)^{1/3}$ and $n^* = n_d + \frac{11}{32}n \simeq \frac{11}{32}n$, with n being the carrier density ($\epsilon = 4\pi\varepsilon_0\varepsilon$, with $\varepsilon = 12.88$ for GaAs) [52,99,100]. Although Landau level mixing is thought to be important in reducing the excitation gaps, the broadening of the Landau levels by the isotopic disorder is well captured by including only the finite width of the wave function [53].

B. Electron-phonon renormalization of the bands and the isotopic shift

The 0 K electron-phonon renormalization is −90 meV for the lowest direct gap E_0 of GaAs and is mainly responsible for the isotopic shift of the bands [58,101]. More precisely, the difference in the renormalization energy between two isotopic compositions corresponds to an isotopic energy shift of the conduction or valence band. This is due to the fact that the different isotopic masses M lead to different electron-phonon renormalization energies, proportional to $M^{-1/2}$. These isotopic shifts have been measured experimentally for the lowest direct band gap E_0 in GaAs of ∼4.52 K (0.39 meV) per amu [61,101], the lowest indirect gap E_g in Si of ∼12.03 K (1.04 meV) per amu, and many other semiconductor materials [58].

The value of ΔE^{71} corresponding to the conduction band offset in a heterostructure between pure ^{69}GaAs and ^{71}GaAs cannot be directly measured from the band gap energy shift ΔE_g^{71} since both the conduction and valence bands contribute.

However, a method to separate the individual contributions originating from the valence and conduction bands from the total shift of the band gap was discussed in Ref. [71] for Si and is based on calculations of the temperature dependence of electronic band states in Ref. [60]. In the case of Si it was concluded that 75% of the isotopic dependence of the band gap energy results from a shift of the valence band edge, and 25% results from an opposite shift of the conduction band edge [71]. Using the same approach, a rough estimation of the energy shift has been made, leading to ∼50% of the band gap energy shift ($\Delta E^{71} = 0.5\Delta E_g^{71}$), or ∼4.52 K (∼0.39 meV), originating from the conduction band [60,102].

IV. DISCUSSION

The eigenvalues of the full Hamiltonian are computed for 10^5 random configuration of ^{71}Ga isotopes for each Landau level filling factor. Each random distribution of isotopes leads to a new eigenvalue energetically shifted from E_c. The statistical distribution of the calculated eigenvalues for different Landau levels and filling factors is shown in Fig. 1. The energy scale is plotted in kelvins for ease of comparison with experimental excitation gap measurements, where the center 0 K corresponds to the unperturbed Landau level E_c. The calculations were performed using $\Delta E^{71} = 4.52$ K for selected filling factors. The lowest Landau level is shown in Figs. 1(a) and 1(b), and higher Landau levels are shown in Figs. 1(c)–1(h) for two carrier concentrations commonly used in experiments. The broadening due to the isotopic disorder is dependent on the magnetic fields and thus the filling factor, and it is inversely proportional to the magnetic length l [40]. Furthermore, the activation gap decreases with decreasing Coulomb energy $e^2/\epsilon l$, as does the isotopic broadening due to its inverse dependence on the magnetic length l, thus not completely closing the smaller gaps at higher Landau levels.

The full widths at half maximum (FWHMs) of the Landau levels were obtained by Gaussian fitting the distribution curves and are plotted as a function of the magnetic length l in Fig. 2. Both figures indicate that this model leads to a broadening of the Landau level that increases with the magnetic field (or decreasing magnetic length) due to the dependence of the magnetic length l on the magnetic field B. Decreasing the magnetic length reduces the spread of the Landau level wave function. As a result, the effect of the isotopic disorder becomes increasingly significant. This behavior is similar to the case of donors and acceptors in Si, where deeper donors and acceptors with more localized wave functions are more affected by the isotopic randomness than the shallower impurities [68–71].

Furthermore, in order to provide a lower range for the size of the effect, the calculations were repeated using a reduced interaction strength $\Delta E^{71} = 1.81$ K, corresponding to the conduction band contributing only 20% of the total band gap shift between ^{71}GaAs and ^{69}GaAs. The FWHMs of the Landau levels are shown in Fig. 2 for both interaction strengths, where the blue circles correspond to $\Delta E^{71} = 0.5\Delta E_g^{71}$ and the red circles correspond to $\Delta E^{71} = 0.2\Delta E_g^{71}$. If the contribution of the conduction band to the 0 K renormalization of the band gap in GaAs amounts to only 20%, the implication would be that the valence band contributes a much larger portion. In this case, the isotopic broadening effect could be much larger for two-dimensional hole gases due to the much larger (80%) energy offset of the valence band [39].

The isotopic disorder could lead to only a suppressed modification of the electron density because of the incompressible nature of the fractional gaps [3,48,103]. Thus, the effect of the isotopic disorder on the fractional gaps is

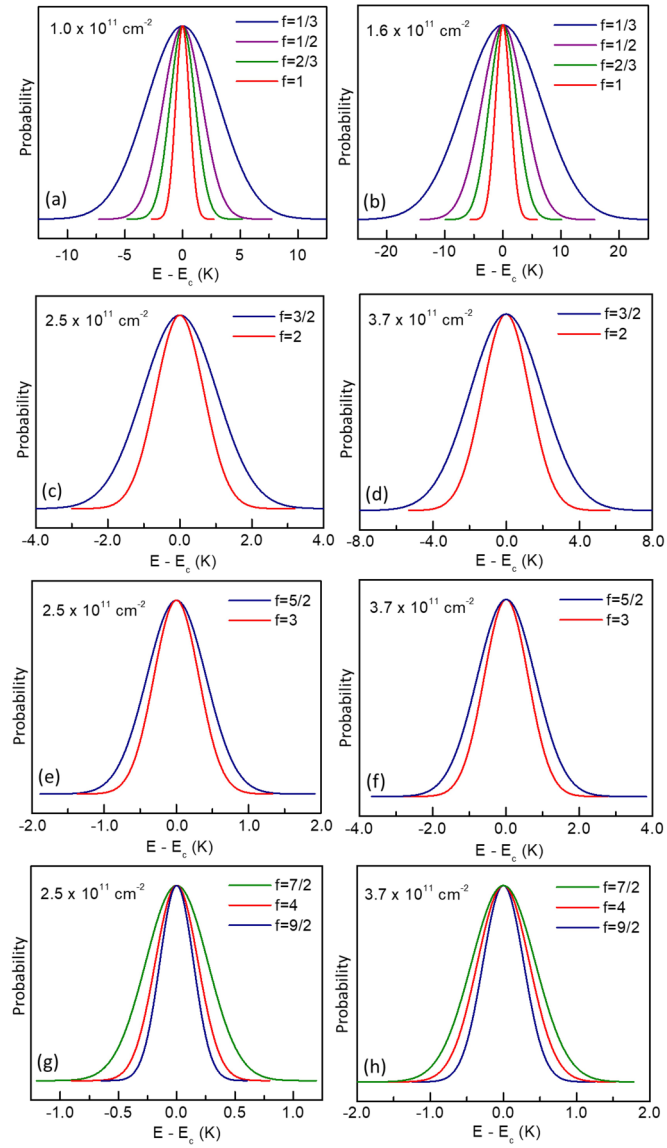

FIG. 1. The statistical distribution of the calculated eigenvalues for (a) and (b) the lowest Landau level and (c)–(h) higher Landau levels, plotted as a function of $E - E_c$. The curves have been normalized for ease of comparison. The energy $E - E_c$ in kelvins corresponds to the energy difference between unperturbed cyclotron frequency $\omega_c = |e|B/m_e$ and the calculated energy. Several carrier concentrations were used which are typically found in experimental measurements. The lowest Landau level distribution at (a) $n = 1.0 \times 10^{11}$ cm^{-2} and (b) $n = 1.6 \times 10^{11}$ cm^{-2} is shown for four different filling factors $f = 1/3, 1/2, 2/3$, and 1. Higher Landau level distributions are shown for two carrier concentrations at (c), (e), and (g) $n = 2.5 \times 10^{11}$ cm^{-2} and (d), (f), and (h) at $n = 3.7 \times 10^{11}$ cm^{-2} for filling fractions $f = 3/2, 2, 5/2, 3, 7/2, 4$, and 9/2.

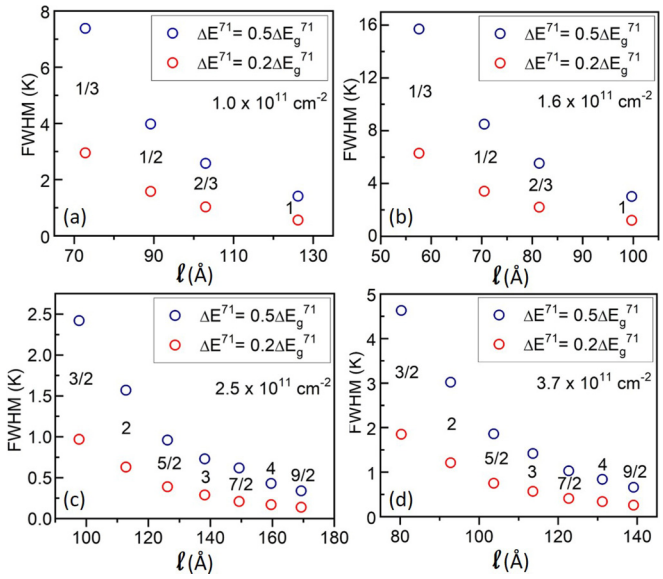

FIG. 2. The full width at half maximum (FWHM) of the distribution of Landau level energies in kelvins, obtained by Gaussian fitting, as a function of the magnetic length l (Å). The calculated Landau level broadening is shown for two conduction band offset values: blue circles show $\Delta E^{71} = 0.5\Delta E_g^{71}$, and red circles show $\Delta E^{71} = 0.2\Delta E_g^{71}$. The lowest Landau level is plotted for two different carrier densities, (a) $n = 1.0 \times 10^{11}$ cm^{-2} and (b) $n = 1.6 \times 10^{11}$ cm^{-2}, and filling factors $f = 1/3, 1/2, 2/3$, and 1, whereas higher Landau levels are shown at (c) $n = 2.5 \times 10^{11}$ cm^{-2} and (d) $n = 3.7 \times 10^{11}$ cm^{-2} for filling fractions $f = 3/2, 2, 5/2, 3, 7/2, 4$, and $9/2$.

difficult to estimate directly, and such calculations would need to include electron-electron interactions. However, temperature-dependent measurements have shown that the fractional gaps close fairly rapidly with increasing temperature, which would indicate that the electron density is affected by electron-phonon interactions [41]. The isotopic disorder should have the same effect as temperature since it affects the phonons through the isotopic mass. Finally, in order to obtain a better understanding of the effect of isotopic disorder, GaAs devices grown with single-isotope ^{69}Ga with isotopic enrichment larger than 99.5% and high chemical purity need to be measured.

V. CONCLUSIONS

In conclusion, the broadening of the Landau levels due to the random distribution of ^{71}Ga and ^{69}Ga isotopes in GaAs was calculated. The isotopes were modeled as short-range δ-function scatterers randomly distributed within the Landau level wave function. The strength of the perturbation is given by the conduction band energy offset between ^{71}GaAs and ^{69}GaAs due to the 0 K renormalization of the conduction band by the electron-phonon interactions. The perturbation by the isotopic disorder leads to a Gaussian distribution of energy eigenvalues for the Landau levels energies. This effect goes beyond two-dimensional electron gases in GaAs and could be even larger for two-dimensional hole gasses. It should occur in silicon devices and is expected to be larger in graphene due to the larger mass difference ratio between ^{12}C and ^{13}C. However, it is particularly interesting in GaAs devices due the smaller gaps of the even-denominator states. The isotopic perturbation and broadening could obscure the underlying physics of the 5/2 state. Removing the isotopic disorder by growing devices using single-isotope ^{69}Ga could help elucidate the physics of existing even-denominator states and possibly reveal new states.

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