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Large splitting of the cyclotron-resonance line in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$/GaN heterostructures

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Cyclotron-resonance (CR) measurements on two-dimensional (2D) electrons in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$/GaN heterojunctions reveal large splittings (up to 2 meV) of the CR line for all investigated densities, n_{2D}, from 1 to $4\times10^{12}\ \mathrm{cm}^{-2}$ over wide ranges of magnetic field. The features resemble a level anticrossing and imply a strong interaction with an unknown excitation of the solid. The critical energy of the splitting varies from 5 to 12 meV and as $\sqrt{n_{2D}}$. The phenomenon resembles data from AlGaAs/GaAs whose origin remains unresolved. It highlights a lack of basic understanding of a very elementary resonance in solids.

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Ever since its first demonstration in 1953,[1] cyclotron resonance (CR) has become the most widely used technique to determine effective masses of carriers in semiconductors and their heterojunctions. In the presence of an external magnetic field, B, electrons of charge e are set into cyclotron motion whose frequency is given by $\omega_c = eB/m^*c$. An rf or far-infrared source is swept which generates a single absorption line at ω_c, from which the effective mass m^* is deduced. CR has also been instrumental in detecting other excitations of the solid via the excitations interaction with CR. They often take on the shape of a level anticrossing from which the energy of the new excitation and its coupling to CR can be inferred. These are well-established, textbook phenomena, which make CR one of the best-understood tools in solid-state research.[2] Yet, in our experiments on the comparatively new $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$/GaN heterostructures we observe huge splittings in the CR line, indicating a strong interaction with an excitation whose origin remains unknown. Our results provide a fundamental challenge to our apparently thorough understanding of CR in solids.

For $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$/GaN heterostructures there exists only a small amount of CR data.[3–6] Our study of CR in two-dimensional electron systems (2DESs) in a sequence of high-quality $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$/GaN structures with a wide range of carrier densities (1–$4\times10^{12}\ \mathrm{cm}^{-2}$) reveals splittings in the CR line reminiscent of level anticrossings, reaching 20% of the resonance energy. While their origin is not established, they resemble splittings seen previously in the CR of AlGaAs/GaAs and Si systems. Our data do not support a universal *ad hoc* model put forward in the AlGaAs/GaAs work and point to the lack of a theoretical understanding of this phenomenon, now seen in three different two-dimensional (2D) systems.

Our $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$/GaN samples are grown by plasma-assisted molecular-beam epitaxy (MBE) on thick GaN ($\sim$15 μm) templates prepared by hydride vapor phase epitaxy (HVPE) on the [0001] face of sapphire. The use of thick HVPE-grown templates is essential for achieving low threading dislocation densities in the GaN buffer region. The reduction of the threading dislocation density has been shown to improve low-temperature mobility at low-electron densities. The typical MBE layer sequence consists of 400 nm of GaN, followed by a layer of 25 to 50 nm $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$, which is capped by 3 nm of GaN. In contrast to the AlGaAs/GaAs system, nitride heterostructures are not modulation doped. The creation of a 2DES rather originates from strong spontaneous and piezoelectric fields arising at the heterointerface.[7,8] The Al mole fraction and the thickness of the barrier layer control the 2D electron density. In our samples the Al content varies from 3% to 10%, yielding the parameters listed in Table I. Samples with $n_{2D}\sim 1-3\times10^{12}\ \mathrm{cm}^{-2}$ show the integer[9] and the fractional quantum Hall effects,[10] further attesting to the high quality of the material.

All cyclotron-resonance measurements were performed at 4.2 K. A Fourier transform spectrometer with light pipe optics was used in combination with a composite Si bolometer for the detection of the far-infrared magnetotransmission. The backsides of samples 3, 5, 8, and 10 were wedged to $\sim 9°$ to reduce Fabry-Pérot interferences. The B field was applied normal to the 2D layer and the density of the 2DES was determined *in situ* via the Shubnikov–de Haas effect. The electron densities in samples 1–6 could be persistently increased (up to 30%) through illuminations with a blue light-emitting diode for 20–60 s.

Figure 1(a) shows a representative set of transmission spectra for sample 3 in different magnetic fields. All spectra

TABLE I. Parameters of all $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$/GaN 2DES samples used in our experiments. n_{2D} is in units of $10^{11}\ \mathrm{cm}^{-2}$ and the mobility, μ, in $10^3\ \mathrm{cm}^2/\mathrm{Vsec}$. E_{crit} and ΔE are in meV. The values in parentheses are estimated from deconvolutions.

Number	n_{2D}	μ	E_{crit}	ΔE	Number	n_{2D}	μ	E_{crit}	ΔE
1	9.8	16	4.9	1.6	6	12.6	17	6.5	1.3
2	11.2	15	5.6	1.7	7	19.0	16	7.3	(0.8)
3	11.4	16	5.6	1.2	8	23.0	18	8.9	1.7
4	11.9	20	5.8	1.7	9	35.8	8	10.4	(1.6)
5	12.3	18	6.1	1.4	10	36.1	19	12.2	(2.0)

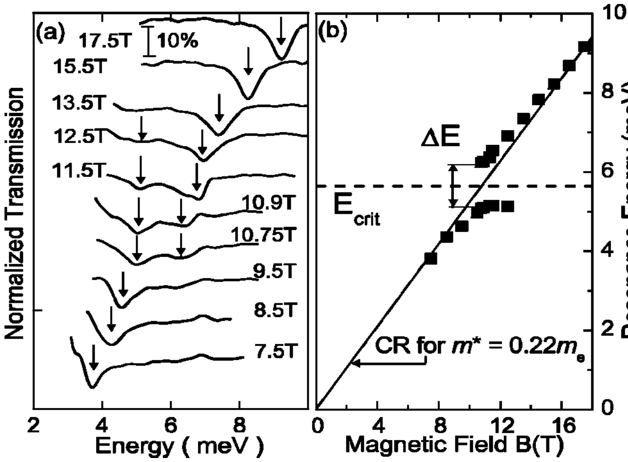

FIG. 1. (a) Far-infrared transmission data on a 2DES of density 1.14×10^{12} cm^{-2} in Al$_x$Ga$_{1-x}$N/GaN (sample 3) for various magnetic fields, B, normal to the 2DES. All data are normalized to the transmission at $B=0$. Traces are offset vertically for clarity. A 10% transmission loss is indicated as a vertical bar. Sharp CR lines are observed for $B<8$ T and $B>15$ T. In the intermediate-field regime large splittings occur. (b) Peak positions of transmission minima of (a) as a function of magnetic field. High- and low-field data follow the CR with an effective mass of $m^*=0.22m_e$. Around 11 T an apparent level anticrossing occurs with a critical energy $E_{\text{crit}}=5.6$ meV.

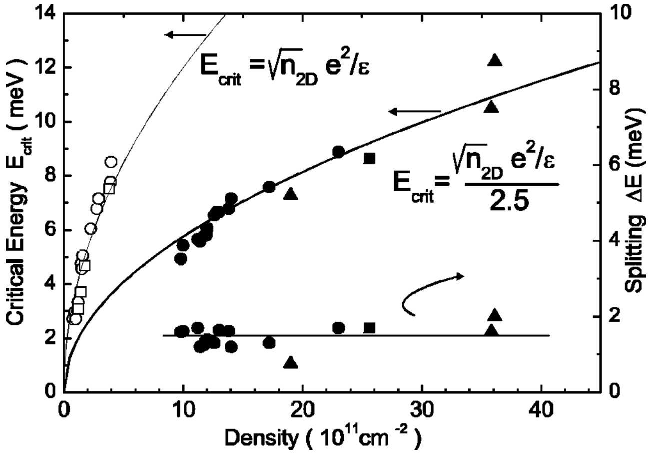

FIG. 2. Critical energy, E_{crit}, (left scale) and splitting, ΔE, (right scale) versus electron density. All Al$_x$Ga$_{1-x}$N/GaN samples of Table I are shown plus data from runs in which the density was increased (up to 30%) by light. Data from clearly resolved CR splittings are indicated by filled circles; broadenings are shown as filled triangles. The full squares represent data from Ref. 3. Open circles and open squares refer to AlGaAs/GaAs data (Ref. 15) and Si data (Refs. 17 and 18), respectively. All data follow a $\sqrt{n_{2D}}$ behavior, however, with very different prefactors. The data set at the bottom (ΔE) shows the magnitude of the CR splitting at B_{crit} as a function of density for all Al$_x$Ga$_{1-x}$N/GaN samples. In the case of line broadening (triangles and square) the "splitting" was estimated from a deconvolution of the broadened CR line.

are taken with 0.24-meV resolution and are normalized to the spectrum obtained at 0 T. For clarity the data are vertically offset. For fields $B>15$ T and $B<8$ T singular sharp resonance dips are observed in transmission. They represent the characteristic CR of the electrons. The 2D nature of the carrier motion is verified by observing the expected $1/\cos\theta$ shift of the resonances under tilted B field. These high- and low-field resonance positions are linearly dependent on B, consistent with each other, and yield an electron effective mass of $m^*=0.22m_e$ which is very close to the literature value.[4]

While the CR is well behaved in the high-field and low-field regimes, at intermediate fields (8 T$<B<15$ T) it deviates considerably from a simple linear relationship. As the field is reduced below $B\sim17$ T the linewidth increases gradually and develops a separate transmission dip near 13 T. For example, at $B=12.5$ T a strong resonance exists near 6.9 meV and a weaker one at 5.1 meV. Lowering the field further, the high-energy resonance gradually broadens and weakens, while the low-energy resonance gains in strength. At $B\sim11.3$ T, both resonances are of equal strength. The integrated intensity of the combined resonances remains comparable to those of the single CR line for $B>15$ T. As B moves towards 8 T, the low-energy resonance continues to grow and develops again into a single CR. For lower fields, the resonance dips regain their sharpness and move linearly with B to smaller energies.

Figure 1(b) shows the position of the resonance energies of sample 3 versus magnetic field. The solid line is a fit to the high- and low-field data. Between 10 T and 14 T we observe a marked deviation from a linear dependence together with the appearance of a second resonance for each magnetic field. The two resonance branches are separated by a gap, ΔE, of ~1.2 meV. The midpoint of this gap, E_{crit}, is ~5.6 meV. The splitting resembles a level anticrossing between the CR of the system and some other resonance at ~5.6 meV. However, this "other resonance" is not observed outside of the crossing regime. In particular, it is not optically active at $B=0$.

A clear splitting in the CR is observed in seven different samples and a distinct line broadening is observed in three others. Figure 2 shows the dependence of E_{crit} and ΔE on the 2D density, n_{2D}, for all ten samples. For specimens in which the splitting is not resolved, we take E_{crit} as the energy of the maximally broadened line. The density dependence of the CR splitting, considering the scatter of the data, is approximately constant, $\Delta E\sim1.5$ meV. Beyond our own data, Wang *et al.*[3] reported a broadening of the transmission dip near 8.6 meV. We obtained this specimen, determined its density, and included these data in our graph. Their position indicates that the same phenomenon is at work. Other CR experiments on Al$_x$Ga$_{1-x}$N/GaN heterostructures do not report a splitting nor a broadening.[4–6] These experiments were performed on low-mobility and very high-density specimens ($\sim3.1\times10^{12}<n_{2D}\lesssim18\times10^{12}$ cm^{-2}). Judging from Fig. 2, the level anticrossing position in most of these experiments exceeded the maximum field employed. In others the splitting may have been missed because of much wider CR linewidths and sparse data sets.

The splitting of the CR line in our samples appears to be the result of the coupling of the 2DES CR with another resonance of the system with an energy in the 5–12-meV range. Interaction between the 2D electrons and bulk or interface phonons cannot be the cause of the CR splitting since all optical-phonon energies in Al$_x$Ga$_{1-x}$N/GaN are above 65 meV.[11,12] The common splitting due to a coincidence of the CR with the intersubband separation of the 2DES can also be ruled out: (i) the calculated subband separation is $\sim$23 meV for $n_{2D} \sim 1 \times 10^{12}$ cm^{-2},[13] which is much bigger than the observed $\sim$6 meV; (ii) such interaction occurs only at finite angles, whereas we observe the splitting for $\theta = 0°$ and find no observable angular dependence. We can also safely rule out a coincidence of opposite spin states in neighboring Landau levels.[14] For our material system such a coincidence would occur for $\theta \sim 77°$. The observed splitting cannot be due to a coupling of the 2DES with any 3D plasmon in any of the layers since there are very few carriers in the bulk. For example, for $n_{2D} \sim 1 \times 10^{12}$ cm^{-2}, the splitting occurs near 6 meV which requires a carrier density of $\sim 10^{17}$ cm^{-3} in a 3D plasmon, whereas *C-V* measurements of the bulk set those densities to $\lesssim 5 \times 10^{14}$ cm^{-3}.[9]

The closest resemblance to our observations is found in a previous report by Schlesinger *et al.*[15] who observed low-energy splittings in the CR of AlGaAs/GaAs heterostructures covering a density range of $1\text{–}4 \times 10^{11}$ cm^{-2}. Their data remain unexplained (see, e.g., Ref. 16). Several observations of Ref. 15 are similar to ours: (i) the CR splitting is not affected by small tilt angles, (ii) no absorption is seen in the $B = 0$ T spectrum at the critical energy, (iii) at the critical B field two resonances of roughly equal width and strength appear, and (iv) the energy, E_{crit}, at which a broadening/splitting is observed is proportional to $\sqrt{n_{2D}}$ (see Fig. 2). While the similarities in these observations suggest a common origin of the CR splittings in AlGaAs/GaAs and our Al$_x$Ga$_{1-x}$N/GaN interface, there are some differences. In AlGaAs/GaAs heterostructures the splitting appears ‘‘abruptly’’ whereas our Al$_x$Ga$_{1-x}$N/GaN data show a gradual evolution of the splitting. Moreover, the separation between the split lines at the critical B field is $\sim$5% of the CR energy in the AlGaAs/GaAs case, much smaller than the $\sim$20% seen in our Al$_x$Ga$_{1-x}$N/GaN heterostructures.

The most important difference is a pronounced deviation of our data from a universality proposed by the authors of Ref. 15. By including previous results of Wilson *et al.*[17] and Kennedy *et al.*[18] on CR line broadening in Si, Schlesinger *et al.* infer that the critical energy follows a universal relationship $E_{\text{crit}} = \sqrt{n_{2D}} e^2/\epsilon$, where ϵ is the dielectric constant. While our data also seem to follow a $\sqrt{n_{2D}}$ dependence, they deviate by a factor of $\sim$2.5 from the AlGaAs/GaAs and Si cases (see Fig. 2). Obvious differences in ϵ and m^* between these materials cannot resolve the discrepancy. In fact, ϵ and m^* of Si and GaN are within 15% of each other and yet there exists a factor-of-2.5 discrepancy in Fig. 2. A plot of E_{crit} vs filling factor, ν, generates no apparent relationship. On the other hand, in a plot of B_{crit} vs n_{2D} the AlGaAs/GaAs and Al$_x$Ga$_{1-x}$N/GaN data fall onto the same line. However, now the Si data deviate by a factor of $\sim$3.

As to the origin of the CR splitting, Schlesinger *et al.* propose an *ad hoc* model. The splitting is conjectured to result from a softening of the large $q \sim 2/l_0$ magnetoroton mode leading to a degeneracy with the CR at $q = 0$. The plasmon wave vector is denoted as q and $l_0 = \sqrt{\hbar c/eB}$ is the magnetic length. Disorder breaks translation invariance, couples both modes, and causes the splitting. For this reason, the splitting is absent in very high-mobility specimens such as AlGaAs/GaAs with $\mu > 10^6$ cm^2/Vsec. The origin of the softening of the magnetoroton and its B dependence remains speculative in Ref. 15. Kallin and Halperin[19] are among the first to address the field dependence of the magnetoplasmon dispersion. Their analysis is limited to high magnetic fields in which the cyclotron energy $\hbar\omega_c \gg E_c$, the Coulomb energy. Although in the experiments $E_c \approx \hbar\omega_c$, the authors provide some general remarks about the CR splitting in AlGaAs/GaAs. According to their calculation the magnetoroton minimum never approaches the CR energy but always exceeds it. Even if the magnetoroton were to cross the CR due to some higher-order interaction it would do so in the wrong direction. The roton minimum is expected to move downward in energy with decreasing field, in contrast to experiment, which requires the minimum to move upward in energy with decreasing field. A calculation by MacDonald,[20] which includes higher-order effects on the magnetoplasmon dispersion, also concludes that the corrections are too small to even bring the minimum into resonance with the CR. A recent calculation on interactions between magnetoplasmons by Cheng[21] asserts that the magnetoroton minimum actually can cross the CR energy. Cheng calculates dispersion relations only in the absence of disorder and does not derive actual splittings of the CR line. Furthermore, as previously argued by Kallin and Halperin, the direction of crossing of the magnetoroton minimum, calculated by Cheng including these higher-order effects, is again inconsistent with experiment (upward vs downward in energy as a function of B). All previous references restrict their calculations to integral values of filling factor, ν. Oji and MacDonald[22] consider the case of arbitrary filling factors but find no particularly strong dependence of the roton minimum energy on ν. Instead, they propose that a combined action of spin-density mode (largely below CR) and charge-density mode (largely above CR) may be responsible for the CR line splitting. However, the couplings of spin and charge to the CR mode are very different in strength and run counter to the observation of a simple level anticrossing behavior. There are other attempts at interpreting the AlGaAs/GaAs CR data using a memory function approach. Gold's calculations,[23] performed in the small-q limit, can only account for a broadening but not for a splitting of the CR line. The calculations by Hu and O'Connell,[24] also based on memory functions, remarkably, generate such a splitting. However, the center of gravity of the combined lines (see Fig. 3 of Ref. 24) always tends to reside above $\hbar\omega_c$ in conflict with experiment. Furthermore, extrapolating these results, equal amplitude of the peaks would occur at $B \sim 8$ T ($E_{\text{crit}} \sim 5.3$ meV) which deviates considerably from the $E_{\text{crit}} \sim 12$ meV of Ref. 15.

At this stage, we do not understand the splitting in the CR of 2D electrons in Al$_x$Ga$_{1-x}$N/GaN heterostructures. The splitting is enormous, reaching up to $\sim$20% of the CR energy. Its unambiguous observation in a second material system establishes the splitting as a general 2D phenomenon rather than being peculiar to just AlGaAs/GaAs.

Theoretical models that address the splitting in AlGaAs/GaAs appeal to a softening of the large wave-vector magnetoplasmon mode, which mixes into the CR due to disorder. Yet the origin, *B*-field dependence, filling factor dependence, magnitude, and even the exact mode responsible for the mixing remain unresolved. At the present level of understanding of 2D electron dynamics it is remarkable that such a huge effect on one of the most fundamental excitations of a solid, the cyclotron resonance of electrons, remains obscure. Our data on the new $Al_xGa_{1-x}N/GaN$ heterostructures clearly highlight the need for a detailed evaluation of the magnetoplasmon mode. Our observed density dependence, which differs significantly from an earlier conjecture, should provide a means to differentiate between various theoretical models.

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