Artificially Constructed Plasmarons and Plasmon-Exciton Molecules in 2D Metals

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Resonant optical excitation was used to create a macroscopic nonequilibrium ensemble of "dark" excitons with an unprecedented long lifetime in a two-dimensional electron system placed in a quantizing magnetic field. Exotic three-particle and four-particle states, plasmarons and plasmon-exciton molecules, coupled with the surrounding electrons through the collective plasma oscillations are engineered. Plasmarons and plasmon-exciton molecules are manifested as new features in the recombination spectra of nonequilibrium systems.

Convincing evidence has been acquired regarding the existence of three-particle excitations participating in collective plasma oscillations in metals, i.e., plasmarons. Plasmarons were predicted as far back as in 1967 [1]. However, the first unambiguous demonstration of their existence has been presented only recently. Plasmaron peaks were observed near the Dirac point in the angle-resolved photoemission spectra of graphene [2]. Plasmaron features were also seen in GaAs/AlGaAs quantum wells (QWs) by means of pulsed tunneling spectroscopy [3]. There have also been a few reports of plasmarons in 3D metals [4]. The plasmaron is essentially a specific example of a more general class of charged excitations in metals, trions, which have been extensively discussed with respect to recombination spectra of two-dimensional (2D) electron systems [5,6]. The main issue of trion physics is related to the possibility of observing the recombination of free (translation invariant) trions [7]. In the work presented here, we show that a trion, a plasmaron, as well as a more complex four-particle plasmon-exciton molecule, form naturally in a 2D metal placed in a quantizing magnetic field.

Before discussing the possibility of observing 2D plasmarons, it is necessary to state the problem of creating 2D translation-invariant two-particle exciton states in metals. These states are likely to form near the Dirac point in graphene owing to a significant reduction of the electron-hole masses [2]. In the case of massive systems, the formation of exciton states becomes more intricate owing to screening of the excitons by free carriers [8] or localization of excitons in a random potential [6]. Even if this is achieved, a question arises whether the third particle attached to the two-particle state would manifest itself in the optical spectra. A photon is unable to create a charged three-particle excitation, and a translationary invariant three-particle excitation cannot decay with photon emission. The influence of the third particle can be visualized through inner transitions of a three-particle state only [7]. Thus, to observe plasmaron features in optical spectra, one has to create a system of free excitons with a lifetime long enough to catch an extra particle. Then a photon emitting transition of the three-particle state should occur. Finally, the emitted photon should fall in a frequency range accessible for its reliable detection. It turns out that all those conditions are well fulfilled for a high mobility electron gas confined in a GaAs/AlGaAs quantum well.

As was already mentioned, the excitons are unstable in metals, such as 2D-electron gas [8]. However, in quantizing magnetic fields and at temperatures well below the cyclotron quantization energy, a 2D metal turns into a special type of 2D insulator at even integer quantum Hall states [9]. Excitations in quantum Hall insulators are magnetoexcitons. They combine properties of excitons in 2D insulators and plasmons in 2D metals [10]. The simplest realizations are magnetoexcitons formed by an electron promoted from the occupied zeroth Landau level to the empty first Landau level and by the vacancy left in the Fermi sea of electrons. There are two magnetoexcitons: a spin singlet with total spin $S = 0$; and a spin triplet with total spin $S = 1$ and spin projections along the magnetic field axis $S_z = -1, 0, 1$ [11]. The singlet magnetoexciton $S = 0$ is essentially the Kohn magnetoplasmon [12]. Its fast relaxation to the ground state occurs via dipole cyclotron radiation [13]. In contrast to the spin singlet, the spin-triplet magnetoexciton is not radiatively active owing to electron spin conservation. It is a "dark" magnetoexciton. It interacts with all other electrons below the Fermi level. The many-body Coulomb interaction lowers its energy below the cyclotron energy. Thus, the spin-triplet magnetoexciton is the lowest energy excited state in the system [11]. Because of this and the spin conservation, the spin-triplet magnetoexcitons exhibit

extremely slow relaxation with the relaxation time reaching hundreds of microseconds [14].

An experimental technique for the creation, manipulation, and detection of macroscopic ensembles of spin-triplet magnetoexcitons has been recently developed [14]. The ensemble can be created with a resonant optical excitation (the resonances appear due to the orbital and the spin quantization), whereas the usage of extra nonresonant photopumping adds high-energy electron-hole pairs to the ensemble. During relaxation, the latter may dissociate and relax to the ground state. Yet, depending on the amount of extra high-energy pairs and the density of spin-triplet magnetoexcitons, some of the dissociated valence holes stick to the magnetoexcitons as extra particles with the formation of positively charged three-particle states, whereas some nondissociated pairs stick to the magneto-excitons with the formation of neutral four-particle states. Negatively charged three-particle states are also likely to form, but since the extra electron settles in the conduction band, they are not optically active in the visible spectral range and therefore are not discussed here. Thus, by manipulating the densities of resonant and nonresonant excitations, one can artificially construct all possible stable states consisting of spin-triplet magnetoexcitons, electrons, and holes.

To create a macroscopic ensemble of spin-triplet excitons, we utilized two sets of high-quality heterostructures with symmetrically doped GaAs/AlGaAs single quantum wells of two widths: 17 and 35 nm (see the Supplemental Material [15]). At the elevated temperatures spin-triplet magnetoexcitons are "ionized" [17], and the recombination spectra exhibit a Zeeman doublet related to the transitions of photoexcited valence holes to the occupied states in the conduction band (Fig. 1).

As the temperature drops, spin-triplet magnetoexcitons are accumulated due to increasing their lifetime [14], and two extra lines emerge (pointed by triangles in Fig. 1). Let us consider feasible optical transitions from the zeroth Landau level of electrons to the zeroth Landau level of heavy holes. The number of such transitions is not large since the spectrum of single-particle states is quantized, and the Fermi level lies in the middle of the cyclotron energy gap separating the zeroth and the first Landau levels. Two obvious transitions are allowed in each of the two polarizations of the emitted photon (Fig. 2). The first transfers the photoexcited valence hole to the conduction band. It is this transition that is observed at elevated temperatures. The second occurs between inner states of a trion formed by a valence hole and a spin-triplet magnetoexciton. This transition is dipole allowed if it does not change the internal quantum numbers of the trion [7]. The final state of the second transition in σ_- polarization is a trion with vacancies in the zeroth Landau level having spins directed oppositely (Pln). In σ_+ polarization, it is a trion with vacancies having aligned spins (T). The excited electron bound in the T state
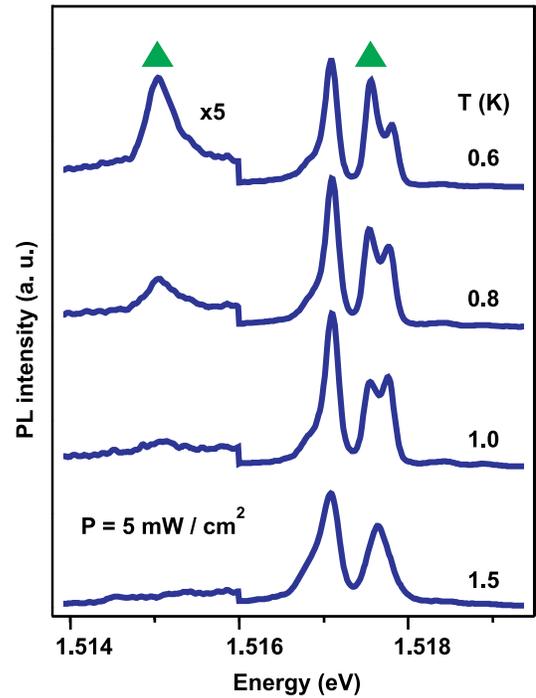
FIG. 1. PL spectra at various temperatures measured at a pumping power density of 5 mW/cm^2.

is not able to jump to any of two available vacancies owing to spin conservation. On the contrary, the excited electron bound in the Pln state may jump to one of two available vacancies with the emission of a cyclotron photon. The latter may in turn be absorbed by the electrons in the zeroth Landau level to give birth to an electron-vacancy pair with the cyclotron energy, and so on. If there was no extra vacancy bound into the spin-triplet magnetoexciton, the described quasiparticle would correspond to a magneto-plasmon with the cyclic quench and the reappearance of electron-hole pairs as plasma oscillations [18]. In the presence of the extra vacancy, the Pln state is a *bihole plasmaron*.

Such sophisticated transitions are clearly observed in the recombination spectra (Fig. 2). The energy of the inner transition for the T trion almost coincides with the energy of single-particle optical transition at elevated temperatures. The tiny energy shift of 0.2 meV is due to the difference in the Coulomb interaction among the particles comprising the T trion as one of the holes passes from the valence to the conductance band. The energy of the inner transition in the Pln trion leading to the appearance of a bihole plasmaron is significantly (2 meV) less than the energy of single-particle optical transition. It reduces so dramatically owing to plasma oscillations.

An exact calculation of the plasmaron energy shift is an exceedingly complex problem [7]. To estimate the plasma oscillation energy, we consider a simplified model of optical transition from the initial two-particle state (magnetoexciton) to the final two-particle state (magnetoplasmon),
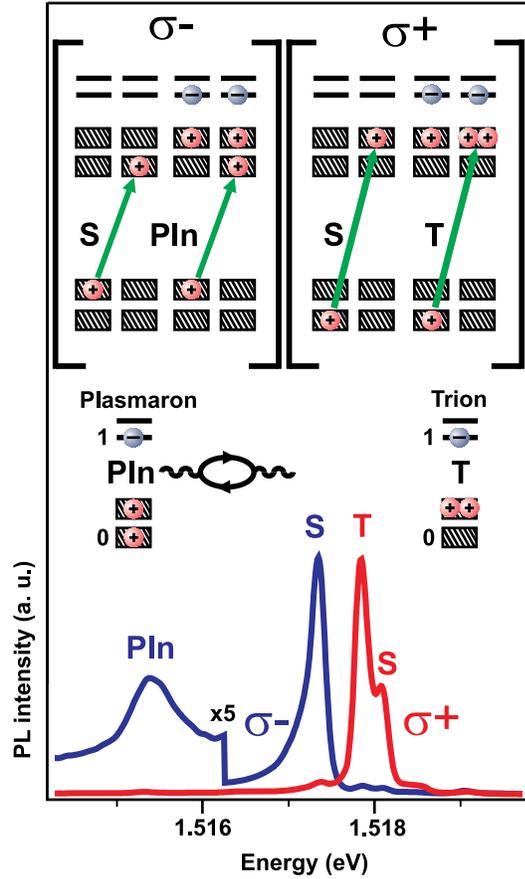
FIG. 2.   Top: Scheme showing allowed optical transitions in σ_- and σ_+ polarizations for single- and three-particle states. The letters S and T (*Pln*) denote the single-particle and trion states, respectively. Bottom: Experimental PL spectra in σ_- (blue) and σ_+ (red) polarizations measured in 35 nm wide QW at 4 T and 0.5 K.

FIG. 3.   Top: PL spectra for narrow QW (17 nm, red curve) and broad QW (35 nm, blue curve). The equally scaled energy axes are shown in the corresponding colors. The energies of single particle optical transitions in σ_- polarization are aligned. The middle inset shows the optical transition scheme used in the theoretical calculations. The top-left inset shows the calculated magnetoexciton (Ex) dispersion curve (red) and magnetoplasmon (MP) dispersion curve (black).

excluding from consideration the vacancy bound in spin-triplet magnetoexciton (Fig. 3). The initial state is a *p*-type exciton (Fig. 3). The minimum of the exciton dispersion is close to momentum $q_m \simeq 1/l_B$ where $l_B = \sqrt{\hbar c / eH}$ is the magnetic length [18]. The contribution to the final state energy made by the plasma oscillations is

$$E_{\mathrm{MP}}(q_m) = \frac{e^2}{\epsilon} q_m f(q_m) e^{-[(q_m l_B)^2/2]},$$

$f(q) = \int e^{-q|z_1 - z_2|} \Psi(z_1)^2 \Psi(z_2)^2 dz_1 dz_2$ is the geometric form factor arising as a result of the nonlocality of the electron in the growth direction of the quantum well (z) (in the strict 2D-limit $f(q) = 1$, see the Supplemental Material [15]). $\Psi(z)$ is the z component of the electron wave function, and ϵ is the relative permittivity of GaAs (Fig. 3). The calculated plasma oscillation energy $E_{\mathrm{MP}}(q_m)$ exceeds the measured plasmaron shift. This disagreement is not surprising considering the second-order corrections to the energies of the quasiparticles in question [11], the

complexity of the real valence hole wave function, and, finally, the influence of the extra vacancy screening the plasma oscillations.

The plasma origin for the *Pln*-energy shift is confirmed by measurements of two quantum wells with significantly different widths [a reduction in the well width enhances the plasma oscillation energy because of the geometrical form-factor $f(q)$] (Fig. 3). In a more narrow QW, the plasmaron shift is larger by 1.4 times, which is exactly in accordance with the calculations. Further, the plasmaron shift measured in a wide range of electron densities agrees well with the calculated values, with the exception of the systematic shift to the lower energies (see the Supplemental Material [15]). Thus, we establish the plasma nature of the energy shift for the *Pln* line. The cross section for the plasmaron recombination increases when either the amount of spin-triplet magnetoexcitons or the number of extra electron-hole pairs increase (Fig. 4). Yet, it saturates at the nonresonant photoexcitation power density of 7.5 mW/cm². With a further increase in the photoexcitation power, the plasmaron recombination quenches and a new recombination channel with an energy shift twice that for plasmarons opens (Fig. 4).

The new recombination line Pl + Ex appears in σ_- polarization of the emitting photon, which signals an optical transition of a hole from the valence band to the lowest spin sublevel of the conduction band (Fig 4). The only possible state one can construct of a spin-triplet exciton and an electron-hole pair apart from the plasmaron is the exotic four-particle state, *plasmon-exciton molecule*. The plasmon-exciton molecule forms at large photoexcitation densities when the probability to meet a photoexcited electron and a photoexcited hole simultaneously in close proximity to a spin-triplet magnetoexciton increases (Fig. 4). The formation of the plasmon-exciton molecule can be visualized as follows. In the initial state, a *p*-type exciton attaches to a spin-triplet exciton forming a biexciton molecule. When the hole bound in the biexciton transfers from the valence band to the conductance band conserving internal quantum numbers of the biexciton, a plasmon-exciton molecule forms. The corresponding recombination line is shifted owing to the plasma oscillations in the final state. In contrast to the plasmaron case, the plasmon-exciton molecule has two electrons both able to jump to the empty vacancy in the lowest spin sublevel (Fig. 4). This doubles the electron charge involved in the collective plasma oscillations. It is surprising that the plasma oscillation energy exactly doubles (within the experimental uncertainty) over the plasmaron energy, notwithstanding the fact that two electrons and only a single vacancy in the electron Fermi sea participate in plasma oscillations (Fig. 4).

In conclusion, we discovered new features in the PL spectrum of the 2D metal which could be clearly attributed to plasmarons and plasmon-exciton molecules because of strong dependence of line intensities on "dark" exciton concentration, energy shift, and photon polarization predicted by theory. The super-long lifetime ensemble of nonequilibrium excitations in 2D metal under a quantizing magnetic field made three- and four-particle stable states possible. Furthermore, these states are heavily connected with collective plasma oscillations. The stability of a bihole plasmaron as well as an exotic four-particle state, i.e., a plasmon-exciton molecule, were demonstrated. The existence of a bound four-particle excited state in metals with collective plasma properties has not been conceived thus far, and hence, extensive theoretical efforts are needed to explain the experimental finding.
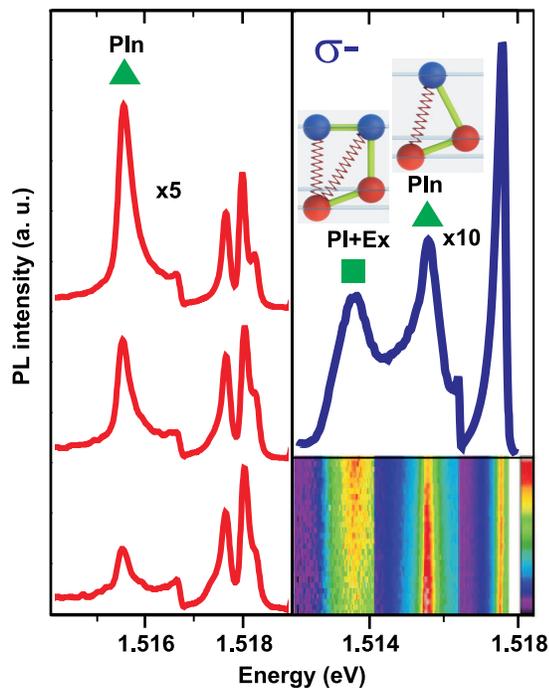
FIG. 4. Left: PL spectra measured at different pumping powers of 1, 3, and 7.5 mW/cm^2, and the bath temperature of 0.5 K (from bottom to top). Right: Bottom, PL spectra measured in pumping power range of 10–80 mW/cm^2 at T = 0.5 K (the power increases in equal steps from bottom to top). Spectra are normalized to give equal intensities for three detected lines at 80 mW/cm^2. Top: PL spectrum in σ_- polarization measured at 80 mW/cm^2. The scheme demonstrates the corresponding final states.

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