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Grating-assisted Raman scattering of plasmons in layered two-dimensional electron systems

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Grating-assisted Raman scattering is introduced as a new mechanism to study the dispersion of near surface modes in dependence of wave vector $K_{\parallel}$ parallel to the surface of a solid. It is shown that a surface grating of a submicrometer period a provides additional transfer of wave vector $K_{\parallel}$ in resonant Raman scattering of plasmons in layered two-dimensional electron systems. Grating-assisted Raman scattering thus allows to extend studies of the two-dimensional plasmon dispersion to a significantly larger range of wave vectors $K_{\parallel}$ than possible by conventional Raman scattering.

The dispersion of collective excitations in layered two-dimensional (2D) electron systems has recently attracted much attention from both theorists[1–3] and experimentalists.[4–7] In systems with a small number N of two-dimensional electron layers, N discrete plasmon branches are expected and experimentally observed. Experimental studies have used resonant Raman scattering in back-scattering geometry. There discrete plasmons with wave vectors $K_{\parallel}$ in the two-dimensional (2D) plane were excited by choosing an appropriate orientation of the 2D system with respect to the incoming and collected radiation. In conventional backscattering geometry, where the incident and scattered light are coupled via the front face of the multilayer sample, the wave vectors of collective excitations are limited to values $K_{\parallel} \leq 4\pi/\lambda_L$ by geometrical considerations alone, where λ_L is the wavelength of the incident radiation. This limits experimental studies of the plasmon dispersion to relatively small wave vectors.

Here we introduce a grating coupler technique in which an additional grating of period a, defined on the surface of the multilayer sample, serves to provide additional momentum transfer in the scattering process. Grating-coupler techniques are well established in absorption studies of surface plasmons in metals at optical frequencies[8] and of 2D plasmons at semiconductor interfaces in the far infrared.[9] In Raman studies of the dispersion of excitations propagating parallel to the surface of a solid we are not aware of any previous use of grating couplers, though gratings have been employed to study and increase coupling of surface-enhanced Raman scattering.[10] We demonstrate that grating-assisted Raman scattering of collective excitations is possible in conventional back-scattering geometry and allows detailed investigation of a large range of plasmon wave vectors up to $(4\pi/\lambda_L)+K_g$, where $K_g = 2\pi/a$ is the grating wave vector. With this new technique plasmon dispersions can be investigated up to significantly larger wave vectors, only limited by the geometry of the grating coupler.

The samples used here are modulation-doped $Al_xGa_{1-x}As/GaAs$ multiple-quantum wells consisting of five 2D electron layers and are from the same wafer (No. 4849) as samples used in the experiments of Ref. 6. The layer sequence in a single period is as follows: 500 Å GaAs, 100 Å $Al_{0.30}Ga_{0.70}As$, 250 Å Si-doped $Al_{0.30}Ga_{0.70}As$, and 50 Å AlAs. On the top of the samples we have prepared grating couplers with different periodicities using holographic lithography. We expose and develop gratings in a thin (100-nm) layer of photoresist. We then evaporate a layer of NiCr under an oblique angle with respect to the surface normal thus obtaining a grating of NiCr stripes. Holographic preparation of photoresist gratings is described in more detail elsewhere.[11]

Measurements are performed at low temperatures with the sample mounted in vacuum on the coldfinger of a closed-cycle helium cryostat. Without illumination sample temperature is typically 11 K. Resonant Raman scattering is performed using an Ar^+ laser pumped tunable dye laser operating around the $E_0+\Delta_0$ band gap of GaAs. The incident radiation is focused by a cylindrical lens producing moderate power densities of about 3 W/cm^2 in the focal line. We use near backscattering geometry as in previous experiments.[6] The scattered radiation is analyzed by a triple Raman spectrometer (Dilor *xy*) with multichannel detector.

The Shubnikov-de Haas oscillations of the magnetoresistance under saturating illumination conditions become visible in high magnetic fields ($B > 2$ T) and exhibit a single period corresponding to a 2D carrier density of $n_s = 7.4 \times 10^{11}$ cm^{-2}. This indicates that the five wells have essentially the same carrier density. The high carrier density makes it likely that more than one subband is occupied though we observe no indication of a second oscillation period in the magnetoresistance.

In Fig. 1(a) we present parallel polarized Raman spectra for a sample without grating coupler and in Fig. 1(b) for a sample with a NiCr grating of period $a = 906$ nm. Spectra obtained in parallel polarization are known to reflect the collective excitations of the electronic system.[12] The in-plane wave vector of the incident light $K_{\parallel L}$ is adjusted by rotating the sample around an axis in the surface plane normal to the directions of incident and collected light. If θ_1 is the angle of incidence and θ_2 the angle of collection relative to the surface normal, $K_{\parallel L}$ is given by $K_{\parallel L} = (2\pi/\lambda_L)(\sin\theta_1 + \sin\theta_2)$. The grating wave vector K_g

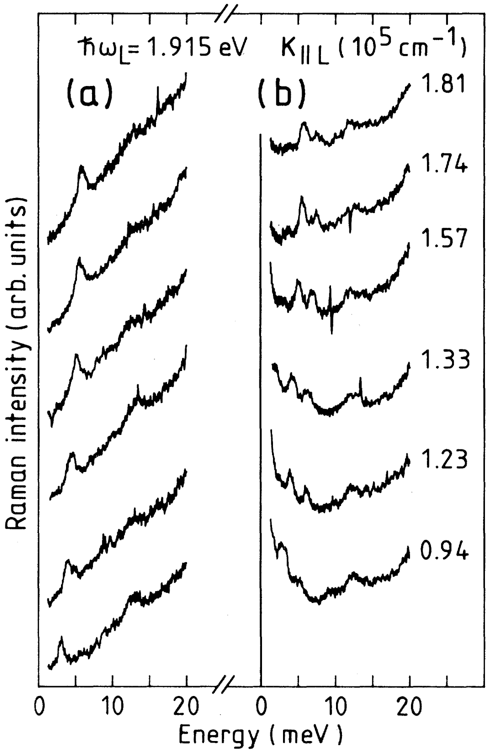

FIG. 1. Parallel polarized spectra for five-layer multiquantum wells (a) without and (b) with a NiCr grating of period $a=906$ nm. The incident laser energy $\hbar\omega_L$ lies close to the $E_0+\Delta_0$ gap of GaAs. Parameter is the wave-vector component $K_{\parallel L}$ of the incident radiation parallel to the sample surface. The sample geometry is discussed in the text.

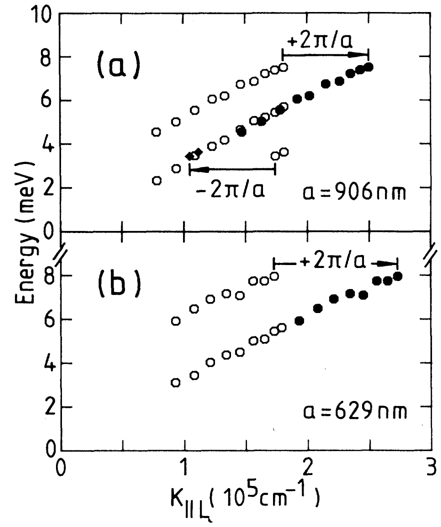

FIG. 2. Dispersion of plasmon modes in five-layer multiquantum wells with grating couplers of period (a) $a=906$ nm and (b) $a=629$ nm. Open dots denote measured values while filled symbols represent values shifted by a reciprocal grating vector $2\pi/a$ as indicated.

lies parallel to $K_{\parallel L}$ and the polarization collected by the spectrometer is perpendicular to the axis of sample rotation. Figure 1(a) shows one sharp plasmon mode below 10 meV and a broad resonance above 10 meV. The energy of the sharp resonance increases with increasing in-plane wave vector. On the sample with grating coupler [Fig. 1(b)] in addition a new resonance of comparable strength at higher energies is observed. This resonance we identify as a plasmon at wave vector $K_{\parallel L}+K_g$, as will be discussed below. The lower energy resonance lies at the same energy position as the one of Fig. 1(a).

In Fig. 2(a) the resonance energy versus in-plane wave vector $K_{\parallel L}$ is shown for the sample of Fig. 1(b). The open dots denote the measured resonance positions. The open dots at higher energies are the grating-coupler-induced resonances. From those we obtain the filled dots by adding a reciprocal grating vector $K_g=2\pi/a$ to the in-plane wave vector $K_{\parallel L}$ as indicated in Fig. 2. This simple addition causes all resonance energies to fall on a single dispersion branch now extending to higher wave vectors. This directly demonstrates that the grating-coupler-induced high-energy resonances are plasmons with in-plane wave vector $K_\parallel = K_{\parallel L}+K_g$. A careful look at the spectra at large $K_{\parallel L}$ [Fig. 1(b)] manifests a weak resonance at lower energies. The resonance positions are also entered in Fig. 2(a). The same arguments as above apply to show that the weak resonances are plasmon modes with smaller wave vector $K_\parallel = K_{\parallel L}-K_g$. At present it is not clear why these small wave-vector modes are much weaker than modes with $K_\parallel = K_{\parallel L}+K_g$. A possible reason for this asymmetry could be blazing effects caused by an asymmetry of the grating profile. For a sample with smaller grating constant of $a=629$ nm a larger wave-vector transfer K_g is observed [Fig. 2(b)]. The low-energy resonance at $K_\parallel = K_{\parallel L}-K_g$ for this sample is probably too weak for detection.

The above conclusions are confirmed by single-particle excitation spectra measured in crossed polarization (not presented here). These spectra are known to show a sharp peak at an energy slightly below $\hbar K_\parallel v_F$ where v_F is the 2D Fermi velocity of the system.[7] The measured spectra for the grating covered samples exhibit an additional broadening or weak shoulders in agreement with the calculated values $\hbar(K_{\parallel L}+K_g)v_F$.

A comparison of this experiment with earlier measurements by Fasol *et al.*[6] on samples from the same wafer is given in Fig. 3. We only show results for the sample with grating period $a=629$ nm and a sample without grating coupler. The data for the sample with $a=906$ nm agree perfectly with the displayed data and are omitted for clarity. Values obtained by grating-induced wave-vector transfer are marked by filled dots, all other values by open dots. Data of Fasol *et al.* are represented by open diamonds. The weak broad structures around 12 meV [Figs. 1(a) and 1(b)] are named A and B and are probably mixed modes of collective intersubband modes I_- (coupled with LO phonons) and the in-plane plasmon modes.[6] Comparison with the experimental results and theoretical dispersions in Ref. 6 suggests that the most prominent plasmon mode observed here corresponds to the $n=5$ multiple-quantum-well plasmon mode where $n=1$ to N is

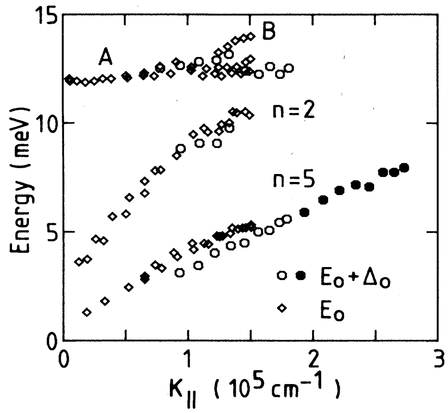

FIG. 3. Comparison of plasmon modes ($n=2$, $n=5$) and coupled modes (A,B) as a function of in-plane wave vector $K_{\parallel}$. Dots represent data measured here and are determined with laser energy $\hbar\omega_L=1.915$ eV near the $E_0+\Delta_0$ gap of GaAs. Open diamonds give data taken from Ref. 6 and are determined with energy $\hbar\omega_L=1.578$ eV near the E_0 gap. Filled dots mark $K_{\parallel}$ values obtained by grating-induced wave-vector transfer.

the mode index also used in Ref. 6. At present, we cannot explain satisfactorily why the $n=5$ branch yields by far the strongest Raman signal in our experiment. Moreover, it is not clear why several plasmon modes of nearly equal strengths are observed in Ref. 6. More recent work by Fasol *et al.*[13] on similar multiple-quantum-well samples shows that occupation of more than one subband gives rise to additional acoustic plasmon modes in which the electrons in different subbands oscillate out of phase. Without a detailed knowledge of the number of occupied subbands in our structure we cannot exclude that the prominent acoustic branch observed here involves electrons in several occupied subbands and is not simply the $n=5$ mode calculated in Ref. 6.

The strength of the grating-induced Raman signal in Fig. 1(b) is nearly the same as the one of the $K_g=0$ signal. This is surprising and indicates that the efficiency of the grating coupler can be rather high. At present it seems very difficult to quantitatively describe this coupling efficiency since the grating geometry is not known in detail and the distance between the grating and the quantum-well layers is of order λ_L/η, where η denotes the refractive index, and not much smaller than the grating period.

In conclusion, we have demonstrated that grating-assisted Raman scattering is a novel tool to study the dispersion relation of near-surface excitations in solids which propagate parallel to the surface. The grating wave vector K_g can be made significantly larger than the in-plane wave vector of the incident radiation. Also, though not yet observed, it should be possible to transfer multiples mK_g ($m=2,3\ldots$) in the Raman process and thus achieve even larger $K_{\parallel}$ transfer. Grating-assisted Raman scattering thus gives access to a much larger range of wave vectors than possible previously and may be used to study new phenomena occurring at large wave vectors such as nonlocal effects in the 2D plasmon dispersion[14] with Raman spectroscopy.

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