Raman spectroscopy of graphite in high magnetic fields: electron-phonon coupling and magnetophonon resonance

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Abstract. The magneto-Raman measurements of graphite were performed in a back-scattering Faraday geometry at temperature 10 K in magnetic fields up to 45 T. The experimental data reveal the rich structure of Raman-active excitations dominated by K-point massive electrons. At high magnetic fields the graphite E_{2g} Raman line shows complex multi- component behavior interpreted as magnetophonon resonance coupled electron-phonon modes at graphite's K-point. Also we found the clear signature of the fundamental, strongly dumped, n=0 magnetophonon resonance associated with H point massless holes.

Keywords: Raman scattering, magnetophonon resonance, graphite, high magnetic fields
PACS: 78.30.Na, 78.20.Ls, 63.22.Rc

INTRODUCTION

The problem of electron-phonon coupling in graphene attracted recently significant attention. Electron-phonon coupling in graphene leads to the significant renormalization and broadening of long-wavelength E_{2g} optical phonons, what are predicted to occur through resonant interaction with Landau- quantized massless Dirac-like fermions in single layer graphene (SLG) [1,2] or massive fermions in bilayer graphene (BLG) [3]. In bulk graphite, the electron band structure combines the massless SLG-like holes and massive BLG-like electrons at the H and K points of the Brillouin zone, respectively. Manifestations of the magneto-phonon resonance (MPR) effect in SLG have been obtained in experiments of magneto-Raman scattering from the surface of bulk graphite and attributed to the presence of graphene flakes decoupled from the bulk graphite [4-8].

RESULTS AND DISCUSSION

In order to probe electron-phonon coupling and inter-Landau level electronic excitations in bulk graphite, we utilized the high magnetic field Raman spectroscopy. The magneto-Raman measurements were performed in a back-scattering Faraday geometry at temperature 10 K in magnetic fields up to 45 T. [9] The experimental data reveal a rich structure of Raman-active excitations dominated by K-point massive electrons. At high magnetic fields the E_{2g} line shows complex multi- component behavior [9]. We interpret different observed modes as MPR coupled electron-phonon modes at graphite's K-point. Also we found a clear signature of the fundamental, strongly dumped, n=0 MPR associated with H point massless holes. In the highest field range (B>35 T) the G line of bulk graphite narrows due to suppression of electron-phonon interaction.

Our results indicate on the necessity to use a full (Slonczewski –Weiss-McClure, SWM [10-12]) band structure model to describe high-field magneto-Raman results. It allowed us to extract accurately the values of SWM band parameters as well as electron-phonon coupling parameters at K- and H-points. In particular, the position and linewidth of unperturbed phonons can be derived from the high-field (> 35 T) spectra, i.e., the position of G-line, 1582**.** 6 cm^{-1}, and the linewidth $FWHM_G$ = 4**.**4 cm^{-1} . We calculated the energies of asymmetric inter-Landau level (LL) transitions using SWM

The Physics of Semiconductors
AIP Conf. Proc. 1566, 165-166 (2013); doi: 10.1063/1.4848337

model. In result, for coupling constants γ, g, and λ [13,14] we obtained γ_r = 44 ±6 cm^{-1}, $g^{(K)}$ = 0.72 ±0.03 cm^{-1}/ T, and extracted $\lambda_r^{(K)}$ ≈3. 3×10^{-2}, in agreement with the values of these parameters previously derived from density functional theory [14]. From analyses of the B -induced modulation of the central component of the G Raman peak near 32T [9], we concluded that this is consistent with MPR due to H -point inter-LL transitions, (1, 0) or (1,− 1), assuming that the LL widths are larger than the coupling strength. At the high-fields (B > 30 T), the MPR-caused changes of G-line position fit well with the model when γ_r= 100±10 cm^{-1} , $g_0^{(H)}$ ~ 3.2 ± 0. 2 cm^{-1}/ $T^{1/2}$, and $\lambda_r^{(H)}$ ≈1. 6 $\times10^{-3}$. It is worth to point that $\lambda_r^{(H)}$ [9] is almost 20 times smaller than $\lambda_r^{(K)}$.

ACKNOWLEDGMENTS

This work was supported by NHMFL UCGP-5068 and DOE/BES DE-FG02-07ER46451. J.K. acknowledges support from DOE/BES through Grant No. DE-FG02-06ER46308 and the Robert A. Welch Foundation through Grant No. C-1509. Y.M. and A.I. acknowledge support from the Texas NHARP Grant No. 01889 and A. P. Sloan Foundation. A.C.F. acknowledges funding from EPSRC Grants No. GR/S97613/01 and No. EP/E500935/1, ERC grant NANOPOTS, EU-FP7 Grants GENIUS and RODIN, and a Royal SocietyWolfson Research Merit Award. The measurements were carried out at the National High Magnetic Field Laboratory, which is supported by NSF Cooperative Agreement No. DMR-0654118, by the State of Florida, and by the DOE.

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