Modulation Spectroscopy and Dynamics of Double Occupancies in a Fermionic Mott Insulator

Rajdeep Sensarma, David Pekker, Mikhail D. Lukin, and Eugene Demler

Physics Department, Harvard University, Cambridge, Massachusetts 02138, USA

(Received 15 February 2009; revised manuscript received 6 March 2009; published 17 July 2009)

Observing antiferromagnetic correlations in ultracold fermions on optical lattices is an important step towards quantum simulation of the repulsive Hubbard model. We show that optical lattice modulation spectroscopy can be used to detect antiferromagnetic order and probe the nature of quasiparticle excitations in a fermionic Mott insulator. At high temperatures, the rate of creation of double occupancies shows a broad peak at frequency of the on-site repulsion U, reflecting the incoherent nature of the hole excitations. At low temperatures, antiferromagnetic order leads to fine structure in the response consisting of a sharp absorption edge reflecting coherent propagation of holes and oscillations as a function of modulation frequency representing spin-wave shake-off processes.

DOI: 10.1103/PhysRevLett.103.035303 PACS numbers: 67.85.−d, 05.30.Fk, 37.10.Jk, 71.10.Fd

Advances in experiments with cold atoms on optical lattices have made them promising candidates for quantum simulators [1,2]. The inherent advantages of cold-atom systems for quantum simulators include the precise tunability of model parameters such as interactions and disorder over wide ranges, e.g., from noninteracting to strongly interacting and completely clean to strongly disordered. Tremendous progress has been made in implementing various quantum Hamiltonians [2] which act as paradigms of strongly interacting systems. These experiments are expected to shed light on long-standing problems in condensed matter physics like existence of spin liquids [3] and the nature of high temperature superconductivity [3,4]. Currently, there is a major effort underway to obtain the antiferromagnetic (AF) fermionic Mott insulator, which is the parent state of many of these exotic phases.

The recent progress in implementing the repulsive Fermi-Hubbard model [5,6] has led to the observation of the Mott insulating state in the large U—strong on-site interaction—limit [7]. Separately, effects of superexchange interactions, which drive antiferromagnetism, have been observed in double-well systems [8]. However, long range AF order is yet to be observed.

Beyond implementing the relevant model, a successful quantum simulator must also be able to probe the properties (e.g., correlation functions) of the resulting state and detect new phases. The usual repertoire of condensed matter experimental probes is not available in the cold-atom setting, and new tools must be developed.

The Mott insulating state at high temperature has been experimentally probed by measuring the production of double occupancies in response to optical lattice modulations [5]. In this Letter, motivated by these experiments, we give a theory of the response of a Mott insulator near half-filling to modulation of optical lattice potential by relating the rate of production of double occupancies to the convolution of spectral function of holes and double occupancies (doublons) in the Mott insulator. We will show that this technique can be used to detect the presence of antiferromagnetic (AF) order and probe the nature of quasiparticle excitations (coherent vs incoherent) in the system. We also discuss the connection of this response to optical conductivity in corresponding charged systems.

We focus on two temperature regimes: (i) the high temperature limit ($J \ll T \sim t_h \ll U$), where T is the temperature, t_h the tunneling matrix, and $J = 4t_h^2/U$ the superexchange scale, which controls the quantum dynamics of the background spins; and (ii) the low temperature limit ($T \ll J$), with an AF ordered spin background.

In the paramagnetic phase (current experiments [5]), we get a response peaked at $\omega = U$ with a width equal to twice the bandwidth of the holes, reflecting the completely incoherent hole and doublon in this limit. We also derive a sum rule for the energy integrated rate of doublon production in this limit.

At low temperatures, AF ordering leads to coherent propagation of quasiparticles and manifests itself in a sharp absorption edge in the production rate. Additional structures at higher energies appear as a result of shake-off processes of spin waves. Thus, lattice modulation spectroscopy can be used to observe the nature of quasiparticle excitations and detect antiferromagnetic ordering in the Mott insulating state.

Modulation of optical lattice.—Repulsive fermions in optical lattices are described by the Hubbard model

$$H = -t_h \sum_{\langle ij \rangle} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \tag{1}$$

where the tunneling matrix t_h and the on-site repulsion U depend (in 3D) on the depth of the optical lattice V through [9] $t_h \sim E_r (\frac{V}{E_r})^{3/4} e^{-2\sqrt{\frac{V}{E_r}}}$ and $U \sim \frac{a_s}{\lambda} E_r (\frac{V}{E_r})^{3/4}$, where E_R is the recoil energy of the photon, λ is its wavelength, and a_s is the s-wave scattering length of the atoms.

The modulation of the optical potential $V(t) = V_0 + \delta V \sin(\omega t)$ effects both t_h and U. The modulation of U can be neglected in the large U/t_h limit. The modulation of $t_h(t) = t_h + \delta t_h \sin(\omega t)$ is related to $V(t)$ by

$$\delta t_h = t_h \delta V\left[\frac{3}{4V_0} - \frac{1}{\sqrt{V_0 E_r}}\right]. \quad (2)$$

The single particle spectrum of the Mott insulator is formed of two bands: (a) the lower Hubbard band, which does not contain double occupancies, and (b) the upper Hubbard band, which contains a single double occupancy. These are separated by the Mott gap $\sim U$. So, the modulation of the optical barrier will produce double occupancies once the frequency of modulation exceeds the Mott gap.

Schwinger bosons and slave fermions.—In the Schwinger boson slave fermion representation, we represent the singly occupied sites (spins) by two Schwinger bosons $a_\sigma^\dagger$, the doubly occupied sites by a doublon $d^\dagger$ and the empty sites by a holon $h^\dagger$. The doublon and holon are fermions. The original fermion creation operator can be written as $c_{i\sigma}^\dagger = a_{i\sigma}^\dagger h_i + \sigma a_{i-\sigma} d_i^\dagger$ with the local constraint equation $a_{i\sigma}^\dagger a_{i\sigma} + d_i^\dagger d_i + h_i^\dagger h_i = 1$. We define the following operators: $F_{ij}^\dagger = \sum_\sigma a_{i\sigma}^\dagger a_{j\sigma}$ and $A_{ij}^\dagger = \sum_\sigma \sigma a_{i\sigma}^\dagger a_{j-\sigma}^\dagger$ which represent the hopping of the bosons and the creation of singlet configurations. We further make the following transformation: On B sublattice, $d^\dagger \to -d^\dagger$. Then the unperturbed Hamiltonian is

$$H_0 = t_h \sum_{\langle ij\rangle}(d_i^\dagger d_j + h_i^\dagger h_j)F_{ij} + t_h \sum_{\langle ij\rangle}(d_i^\dagger h_j^\dagger A_{ij} + \text{H.c.}) + U\sum_i d_i^\dagger d_i, \quad (3)$$

while the perturbation due to the lattice modulation is

$$H_1(t) = \delta t_h \sin[\omega t]\sum_{\langle ij\rangle} d_i^\dagger h_j^\dagger A_{ij} + h_i d_j A_{ij}^\dagger, \quad (4)$$

where we have neglected terms that do not create or destroy doublons. We work with a system at half-filling, at temperatures $T \ll U$, where we can neglect the presence of doublons and holes in the unperturbed system. We also assume that the doublons are created by the action of the perturbation Hamiltonian only; i.e., we neglect the decay of doublons during the time evolution. This approximation is justified as long as $T \ll U$, as the decay of a doublon in the system is a very slow process [10]. Under these assumptions, the rate of creation of doublons (up to 2nd order in perturbation theory), is given by

$$P_d(\omega) = \frac{\pi}{2}(\delta t_h)^2 \int d\omega_1 \int d\omega_2 \sum_{\langle ij\rangle\langle lm\rangle} \mathcal{A}^s_{ijlm}(\omega_2)\mathcal{A}^d_{il}(\omega_1) \times \mathcal{A}^h_{jm}(\omega - \omega_1 - \omega_2), \quad (5)$$

where ω is the frequency of the perturbation, $\mathcal{A}^{d(h)}$ is the spectral function for the doublon (hole), and $\mathcal{A}^s$ is the Fourier transform of $\langle A_{lm}(t)A_{ij}^\dagger(t')\rangle$.

We emphasize that the response we are calculating is not equivalent to optical conductivity in condensed matter systems. (i) The current vertex in optical conductivity is replaced by the kinetic energy vertex. (ii) The optical conductivity involves the convolution $\mathcal{A}^d \circ \mathcal{A}^d$ and $\mathcal{A}^h \circ \mathcal{A}^h$, whereas the calculated response only involves the convolution $\mathcal{A}^d \circ \mathcal{A}^h$. Since the doublon spectral function is shifted by U, as we move away from half-filling, there is no response at low frequencies, whereas there would be optical response in a compressible state.

High temperature.—We now focus on the regime $U \gg T \sim t_h \gg J$, which is the regime of interest for the current experiments. In this limit, the quantum dynamics of the spins are irrelevant, and one can replace the A operators by the probability of finding a ↑↓ or ↓↑ configuration in an ensemble where all spin configurations occur with equal weight. Thus,

$$P_d(\omega) = \frac{\pi \delta t_h^2}{2} \sum_{\langle ij\rangle\langle lm\rangle} P_s \int d\tilde{\omega} \mathcal{A}^d_{il}(\tilde{\omega})\mathcal{A}^h_{jm}(\omega - \tilde{\omega}), \quad (6)$$

where P_s is the probability of finding relevant configurations at (i, j) and (l, m) given by $P_s = 1/2$ if $(i, j) = (l, m)$, $P_s = 1/8$ if (i, j) and (l, m) have no overlap, and $P_s = 1/4$ if (i, j) and (l, m) share one site in common. Because of particle-hole symmetry of the problem at half-filing, we have $\mathcal{A}^d(\omega + U) = \mathcal{A}^h(\omega)$; therefore, it is enough to compute the spectral function for the holes only.

We now try to evaluate the spectral function of a single hole in a spin disordered background. The completely incoherent spectral function of the hole in this limit has been worked out by Brinkman and Rice [11] and Kane *et al.* [12] using the retraceable path approximation.

The Green's function has contributions from processes where the hole hops from one point to another. As the hole hops, it scrambles the spin configuration, and a string of ferromagnetic bonds is required along the path for the process to contribute. The probability of finding such a string is given by $(1/2)^L$, where L is the length of the path. However, the trajectories where the hole retraces its path do not scramble the background spins and have a weight of 1 as opposed to $(1/2)^L$. They provide the dominant contribution to the density of states at low energies.

Within the retraceable path approximation, which is similar to the method used by Anderson in his original paper on localization physics [13], the spectral function of a hole is given by [11] (z is the lattice coordination number)

$$\mathcal{A}(\omega) = \frac{1}{\pi z t_h}\left[\frac{(5 - 9\omega^2/z^2 t_h^2)^{1/2}}{1 - \omega^2/z^2 t_h^2}\right]. \quad (7)$$

The spectral function is plotted as a function of frequency in Fig. 1(a). The spectrum is incoherent and has a bandwidth of $2\sqrt{z-1}t_h$. The spectral weight decreases monotonically as one goes towards the band edge. The rate of production of doublons, calculated using this spectral function, is plotted in Fig. 1(b). There is a peak at $\omega = U$ with weight up to twice the bandwidth for the holes.

We note here some recent work in the paramagnetic phase using different techniques [14,15].

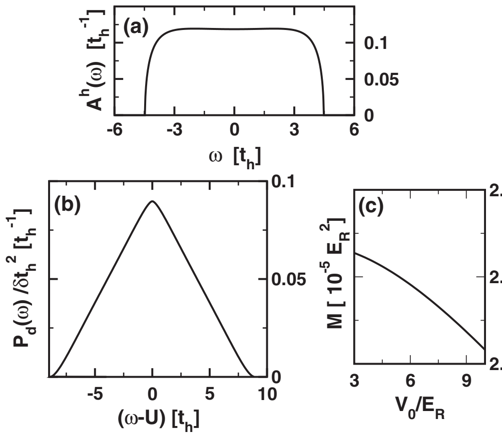

FIG. 1. (a) The density of states of a single hole in a half-filled background in the atomic limit. (b) The rate of production of double occupancies as a function of frequencies and (c) the energy integrated response in high temperature limit.

Sum rule.—In this case, we consider the energy integrated rate of production of doublons $\int d\omega P_d(\omega)$. Using the identity $\int_{-\infty}^{\infty} d\omega \mathcal{A}_{ij}^{d(h)}(\omega) = \delta_{ij}$, we obtain the sum rule in the high temperature limit

$$M = \int_{-\infty}^{\infty} d\omega P_d(\omega) = \frac{\pi}{4}(\delta t_h)^2. \tag{8}$$

The sum rule is proportional to $(\delta t_h)^2$, which is proportional to t_h^2 for a constant fractional change in the amplitude of the lattice potential. Assuming U/t_h is tuned by tuning the lattice potential and a constant fractional change in the amplitude of the potential ($\delta V_0/V_0$ is held fixed), one finds that in the Mott regime, the energy integrated weight monotonically decreases with increase in V_0/E_R, as shown in Fig. 1(c)].

Low temperature (antiferromagnetic phase).—We now consider the response of the system at $T = 0$ in an AF ordered phase. In this regime, the spectral function of holes has a sharp peak and a series of broad features. The sharp peak is reflected in the doublon creation rate as a sharp absorption edge, and the broad features result in oscillations in the rate as a function of modulating frequency.

This phase is characterized by the Bose condensation of $\uparrow$ and $\downarrow$ Bosons on A and B sublattices. In a $1/S$ expansion, the fluctuations are governed by the Holstein-Primakoff Hamiltonian $H = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}}^{\dagger} \alpha_{\mathbf{k}}$, where $\omega_{\mathbf{k}} = zJ(1 - \gamma_{\mathbf{k}}^2)^{1/2}$ is the dispersion of the spin wave with $\gamma_{\mathbf{k}} = (1/3)[\cos k_x + \cos k_y + \cos k_z]$, and the quasiparticle operators are given by $\alpha_{\mathbf{k}} = u_{\mathbf{k}} a_{\mathbf{k}} - v_{\mathbf{k}} a_{-\mathbf{k}}^{\dagger}$ with $a_{\mathbf{k}} = \sum_{i\in A} a_{i\downarrow} e^{i\mathbf{k}\cdot\mathbf{r}_i} + \sum_{j\in B} a_{j\uparrow} e^{i\mathbf{k}\cdot\mathbf{r}_j}$. The coherence factors $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ are given by $u_{\mathbf{k}} = (1/\sqrt{2})(\omega_{\mathbf{k}}^{-1} + 1)^{1/2}$ and $v_{\mathbf{k}} = -\mathrm{sgn}(\gamma_{\mathbf{k}})(1/\sqrt{2})(\omega_{\mathbf{k}}^{-1} - 1)^{1/2}$. The hole hopping Hamiltonian can be written as $H^h = t_h \sum_{\langle ij \rangle} h_i^{\dagger} h_j a_{i\sigma} a_{j\sigma}^{\dagger}$. Replacing the $\uparrow$ and $\downarrow$ spins on A and B sublattice by the condensate amplitude $\sqrt{\rho_0} = 1$, this term can be written as

$$H^h = z t_h \sqrt{\rho_0} \sum_{\mathbf{kq}} h_{\mathbf{k}}^{\dagger} h_{\mathbf{k}-\mathbf{q}} (u_{\mathbf{q}} \gamma_{\mathbf{k}-\mathbf{q}} \alpha_{\mathbf{q}} + v_{\mathbf{q}} \gamma_{\mathbf{k}} \alpha_{-\mathbf{q}}^{\dagger}) + \text{H.c.} \tag{9}$$

The motion of a hole is thus accompanied by creation of a spin wave. We note that we have neglected beyond near neighbor hopping in this calculation. For cold-atom systems, the separable optical potential leads to no hopping along face or body diagonals, while the next-near-neighbor hopping $t' \sim 0.01t$ or lower in the parameter range of interest. We calculate the self-energy of the hole in a self-consistent Born approximation [16,17] by computing the noncrossing Feynman diagram for the self-energy. This approximation has been extremely successful in capturing the essential physics of holes in 2D antiferromagnets and in comparison to exact diagonalization studies [18]. At $T = 0$, the self-energy is given by

$$\Sigma(\mathbf{k}, \omega) = \sum_{\mathbf{q}} |\Gamma(\mathbf{k}, \mathbf{q})|^2 G(\mathbf{k} - \mathbf{q}, \omega - \omega_{\mathbf{q}}), \tag{10}$$

where the vertex function $\Gamma(\mathbf{k}, \mathbf{q}) = z t_h \sqrt{\rho_0}(u_{\mathbf{q}} \gamma_{\mathbf{k}-\mathbf{q}} + v_{\mathbf{q}} \gamma_{\mathbf{k}})$ and the self-consistency is ensured through

$$G^{-1}(\mathbf{k}, \omega) = \omega - \Sigma(\mathbf{k}, \omega). \tag{11}$$

The spectral weight obtained from the self-consistent solution for $J = 0.2t_h$ is plotted for two different $\mathbf{k}$ values, ([0,0,0] and $[\pi/2, \pi/2, \pi/2]$) in Fig. 2(a). At the lowest energy of propagation of the hole, which occurs at $(\pi/2, \pi/2, \pi/2)$, spin waves cannot be created (at $T = 0$) leading to a large coherent weight which gradually decreases as one moves to the center of the Brillouin zone. The location of the coherent peak disperses as $\sim J\gamma_{\mathbf{k}}^2$, corresponding to second order hopping processes which do not scramble the AF alignment. The presence of the coherent peak is a robust feature which goes beyond the approximation used to calculate it here [12].

Beyond the coherent peak, there are additional broad features at higher energies, whose peak-to-peak distance scales with J. These are generated by spin-wave shake-off processes [19]. The peaks correspond to $2, 4, 6, \ldots$ spin waves and are dominated by spin waves near the Brillouin zone boundary where the flat spin-wave spectrum results in a diverging density of states. This is similar to peaks in the 2-magnon Raman response in antiferromagnetic insulators [20]. These peaks are also present in exact diagonalization studies in 2D [21] and hence are robust features of the system.

In terms of the calculated spectral function, one can calculate the rate of doublon production as

$$P_d(\omega - U) = \frac{\pi}{2}(\delta t_h)^2 \kappa \sum_{\mathbf{k}} \gamma_{\mathbf{k}}^2 \int d\tilde{\omega} \mathcal{A}(\mathbf{k}, \tilde{\omega}) \mathcal{A}(\mathbf{k}, \omega - \tilde{\omega}), \tag{12}$$

where $\kappa = 1 - (1/2z)\sum_{\mathbf{k}} \gamma_{\mathbf{k}}^2/\omega_{\mathbf{k}}$ is a vertex correction which takes care of the singlet spectral function. There is no convolution with spin spectral functions as the longitu-

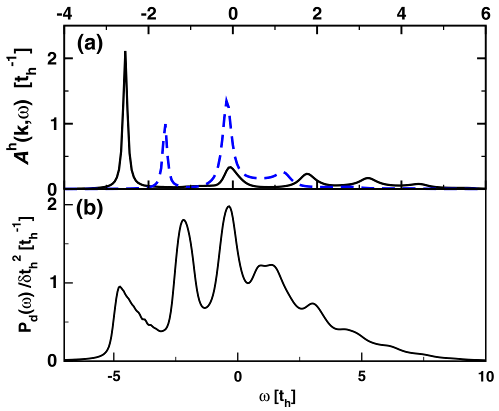

FIG. 2 (color online). (a) The spectral function of the hole at ($\pi/2$, $\pi/2$, $\pi/2$) (solid line) and at (0,0,0) (dashed line) for a system with $U/t = 20$. The spectral functions are broadened by an artificial broadening of $0.06t$. (b) The rate of doublon production $P_d(\omega)$ as a function of perturbing frequency.

dinal modes (↑ on A and ↓ on B) of the condensate are neglected [12].

The rate of doublon production is plotted is Fig. 2(b). It shows an abrupt edge at the lower end of the spectrum corresponding to the coherent spectral weight of the holes. The other oscillations in the response reflect the convolution of the coherent part with the broad incoherent peaks due to shake-off processes and that of the peaks themselves. We thus see that the presence of the AF order leaves its signature in the frequency dependence of the response.

We sketch what happens as we move away from half-filling (still remaining within the AF phase). The order parameter ρ_0 decreases, weakening the scattering of the hole by the spin waves. Thus, the coherent part should grow leading to a sharper edge. This is in contrast to disordering the AF phase by raising temperature, where the scattering from occupied spin-wave modes reduces the coherent part.

We note recent work on response in 1D systems [22].

Comparison with experiments.—In the experiments of Ref. [5], the modulation is kept on for a fixed number of cycles. The time of drive is proportional to ω^{-1}, and the quantity measured is proportional to $P_d(\omega)/\omega$. So the frequency integrated response should decrease even faster with U/t_h as compared to Fig. 1(c). However, the experimental data show a monotonic increase with U/t_h [23].

We believe this could be due to several reasons: (a) The response might be dominated by terms beyond second order perturbation theory. This can be checked by putting on the drive for a different amount of time and looking at the linearity (or lack thereof) of the number of doublons produced with time. (b) The unperturbed system is not in thermal equilibrium due to slow relaxation of the doublons created during tuning of U/t_h [10], or (c) Relaxation of doublons while driving the system leads to a steady state behavior. We hope the discrepancies between the theory and experiments can be settled with further experiments.

Conclusion.—Observing superexchange induced correlations in cold atomic gases on optical lattices is an important step in engineering and simulating strongly correlated phases of matter. In this Letter, we have shown that optical lattice modulation spectroscopy can be used to probe antiferromagnetic order and nature of quasiparticles in the repulsive Fermi-Hubbard model. Further, lattice modulation is a generic technique and can be used to probe other phases like d-wave superfluid or d-density wave [24].

We would like to thank Ehud Altman and Sebastian Huber for useful discussions. This authors acknowledge the support of DARPA, CUA, AFPOSR, and NSF Grant No. DMR-07-05472. R. S. and D. P. were partially supported by NSF Grant No. DMR-05-41988.

[1] R. P. Feynman, Int. J. Theor. Phys. **21**, 467 (1982).
[2] I. Bloch, J. Dalibard, and W. Zwerger, Rev. Mod. Phys. **80**, 885 (2008).
[3] P. A. Lee, N. Nagaosa, and X.-G. Wen, Rev. Mod. Phys. **78**, 17 (2006).
[4] P. W. Anderson, Science **235**, 1196 (1987).
[5] R. Jordens, N. Strohmaier, K. Gunter, H. Moritz, and T. Esslinger, Nature (London) **455**, 204 (2008).
[6] U. Schneider *et al.*, Science **322**, 1520 (2008).
[7] M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. **70**, 1039 (1998).
[8] S. Trotzky *et al.*, Science **319**, 295 (2008).
[9] This relation holds for deep lattices, which is the case for the Mott insulating regime we are interested in.
[10] N. Strohmaier *et al.*, arXiv:0905.2963.
[11] W. F. Brinkman and T. M. Rice, Phys. Rev. B **2**, 1324 (1970).
[12] C. L. Kane, P. A. Lee, and N. Read, Phys. Rev. B **39**, 6880 (1989).
[13] P. W. Anderson, Phys. Rev. **109**, 1492 (1958).
[14] S. D. Huber and A. Ruegg, Phys. Rev. Lett. **102**, 065301 (2009).
[15] C. Kollath, A. Iucci, I. McCulloch, and T. Giamarchi, Phys. Rev. Lett. **97**, 050402 (2006).
[16] F. Marsiglio, A. E. Ruckenstein, S. Schmitt-Rink, and C. M. Verma, Phys. Rev. B **43**, 10882 (1991).
[17] S. Schmitt Rink, C. M. Verma, and A. E. Ruckenstein, Phys. Rev. Lett. **60**, 2793 (1988).
[18] E. Dagotto *et al.*, Phys. Rev. B **41**, 9049 (1990); D. Poilblanc, H. J. Schultz, and T. Ziman, Phys. Rev. B **47**, 3268 (1993).
[19] B. I. Shraiman and E. D. Siggia, Phys. Rev. Lett. **60**, 740 (1988).
[20] A. V. Chubukov and D. M. Frenkel, Phys. Rev. B **52**, 9760 (1995).
[21] P. Beran, D. Poilblanc, and R. B. Laughlin, arXiv:cond-mat/9505085.
[22] F. Massel, M. J. Leskinen, and P. Torma, arXiv:0904.4815.
[23] In the experiments by Esslinger *et al.*, U/t is tuned by tuning both V_0/E_R and a_s/λ. We have calculated the sum rule by using their data points, and it still decreases monotonically.
[24] D. Pekker, R. Sensarma, and E. Demler, arXiv:cond-mat/0906.0931.