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# Sum rules for inelastic light scattering in the Hubbard model

J.K. Freericks[a,*], T.P. Devereaux[b]

[a]Department of Physics, Georgetown University, 37th and O Sts. NW, Washington, DC 20057, USA
[b]Department of Physics, University of Waterloo, Waterloo, ON, Canada N2L 3G1

**Abstract**

The *f*-sum rule for the optical conductivity has been known for many years, and has found widespread use in a number of different fields of science. In this contribution, we show how the *f*-sum rule can be generalized to inelastic light scattering and we explicitly derive the form of the sum rule for the single-band Hubbard model. We discuss evidence that indicates such a sum rule likely holds in already calculated dynamical mean-field theory results and we describe evidence in support of these new sum rules in electronic Raman scattering data for $SmB_6$.


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It has long been thought that neither electronic Raman scattering nor inelastic X-ray scattering have any sum rules, so spectral weight can appear or disappear as a function of temperature. In recent work [1], we showed that there is a sum rule for electronic Raman scattering, which may help with calibration and analysis of experiments and may aid in determining new physics about models that describe real materials.

When we examine the optical conductivity of a solid-state material, and restrict our spectral range to the lowest band, then the *f*-sum rule [2] which counts the total number of electrons in the material, gets replaced by the projected version of the sum rule [3] which measures the average kinetic energy of the material. Hence, using a proper energy cutoff for the optical conductivity sum rule allows one to examine the evolution of the kinetic energy as a function of temperature, pressure, or other parameters. This is a direct way to measure the average kinetic energy of a strongly correlated material. The sum rules for inelastic light scattering are more complicated, and depend on both the potential energy (and for X-rays) the kinetic energy of the material.

Our starting point is to examine the imaginary-time susceptibility of an operator $\mathcal{O}$ that appears in the nonresonant light-scattering correlation function

$$\chi(\tau) = \langle \mathcal{T}_\tau \mathcal{O}(\tau)\mathcal{O}^\dagger(0)\rangle - |\langle \mathcal{O}\rangle|^2, \tag{1}$$

where $\tau$ is the imaginary time operator, $\mathcal{T}_\tau$ is the time-ordering operator, and $\mathcal{O}(\tau) = \exp \tau(\mathcal{H} - \mu\mathcal{N})\mathcal{O} \exp -\tau(\mathcal{H} - \mu\mathcal{N})$ is the imaginary-time-dependent operator. We use the notation $\langle \mathcal{O}\rangle = \mathrm{Tr}\ \exp[-\beta(\mathcal{H} - \mu\mathcal{N})]\mathcal{O}/\mathcal{Z}$ with $\mathcal{H}$ the Hamiltonian, $\mu$ the chemical potential, $\mathcal{N}$ the electron number operator, $\beta = 1/T$ the inverse temperature, and $\mathcal{Z} = \mathrm{Tr}\ \exp[-\beta(\mathcal{H} - \mu\mathcal{N})]$ the partition function. Taking the Fourier transform of Eq. (1) yields

$$\begin{aligned}\chi(\mathrm{i}\nu_l) &= \int_0^\beta \mathrm{d}\tau \mathrm{e}^{\mathrm{i}\nu_l\tau}\chi(\tau) \\ &= \sum_{mn} \frac{\mathrm{e}^{-\beta(E_n-\mu N_n)} - \mathrm{e}^{-\beta(E_m-\mu N_m)}}{\mathrm{i}\nu_l + E_m - \mu N_m - E_n + \mu N_n} \frac{|\langle m|\mathcal{O}|n\rangle|^2}{\mathcal{Z}}\end{aligned} \tag{2}$$

with $\mathrm{i}\nu_l = 2\pi i l T$ the Bosonic Matsubara frequency and we introduced two complete sets of basis states for the Lehmann representation in the last equation ($\mathcal{H}|m\rangle = E_m|m\rangle$ and $\mathcal{N}|m\rangle = N_m|m\rangle$). We will only investigate operators $\mathcal{O}$ that do not change the total electron number, so $N_m = N_n$ throughout.

*Corresponding author. Tel.: +1 202 687 6159; fax: +1 202 687 2087.
*E-mail addresses:* freericks@physics.georgetown.edu (J.K. Freericks), tpd@lorax.uwaterloo.ca (T.P. Devereaux).

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Next, we perform an analytic continuation $i\nu_l \to \nu + i\delta$ and integrate $\nu \operatorname{Im}\chi(\nu)$ from 0 to $\infty$ to get

$$\begin{aligned}\int_0^\infty d\nu \nu \operatorname{Im}\chi(\nu) &= \frac{\pi}{2}\sum_{mn} e^{\beta\mu N_n}(e^{-\beta E_n} - e^{-\beta E_m})(E_m - E_n)\frac{|\langle m|\mathcal{O}|n\rangle|^2}{\mathcal{Z}} \\ &= \frac{\pi}{2}\langle[\mathcal{O},[\mathcal{H},\mathcal{O}^\dagger]]\rangle. \end{aligned} \tag{3}$$

Note that we need the single power of frequency, which yields an even summand over the states $m$ and $n$, because the integral restricts us to $\nu \geqslant 0$. Since the operator $\mathcal{O}$ does not normally commute with either the kinetic energy operator or the potential energy operator in $\mathcal{H}$, the sum rule in Eq. (3) normally depends on both the kinetic and the potential energies (similar ideas were considered by Turlakov and Leggett [4]).

The Hamiltonian we will examine here is that of the Hubbard model [5]

$$\mathcal{H} = -t\sum_{\langle ij\rangle\sigma}(c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma}) + U\sum_i c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}, \tag{4}$$

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) creates (destroys) an electron at lattice site $i$ with spin $\sigma$, $t$ is the hopping integral, $U$ is the screened (local) Coulomb interaction, and the sum over $\langle ij\rangle$ is a sum over nearest-neighbor pairs. We will work on a hypercubic lattice in $d$ dimensions and take the limit $d \to \infty$. Then if we scale the hopping matrix by $t = t^*/2\sqrt{d}$ and use $t^*$ as our energy unit, all many-body correlations become local, and the many-body problem can be solved with dynamical mean-field theory (DMFT) [6].

Inelastic light scattering is a process where light of one frequency is incident on a material; the light then interacts with the charge excitations, and either loses or gains energy before being scattered out of the solid. By examining the number of photons that lose or gain energy as a function of that energy, one can probe the charge excitations of the material. Since light has a polarization vector, controlling the polarization of the incident and reflected photons allows us to project the scattering process onto charge excitations that have a well-defined symmetry in the Brillouin zone (BZ). In the $A_{1g}$ scattering geometry, the incident and scattered light polarizers are parallel, and the photon is sensitive to charge excitations from all regions of the BZ. In the $B_{1g}$ geometry, the polarizers are oriented for example at 45° with respect to the copper–oxygen bonds in a high-temperature superconductor, but are perpendicular to each other. This case probes charge excitations near the BZ edge and away from the BZ diagonal. The full light-scattering process involves two types of terms: terms where the matrix elements for the scattering depend on the incident photon frequency (so-called resonant and mixed scattering terms) and terms that are independent of the incident photon frequency (so-called nonresonant terms). We consider only nonresonant scattering here. The resulting operators for the nonresonant scattering of a photon with wavevector $\mathbf{q}$ can be summarized as follows (in momentum and real space):

$$\begin{aligned}\mathcal{O} &= -\sum_{\mathbf{k}\sigma}\varepsilon(\mathbf{k}+\mathbf{Q})c_{\mathbf{k}+(\mathbf{q}/2)\sigma}^\dagger c_{\mathbf{k}-(\mathbf{q}/2)\sigma} \\ &= \frac{t^*}{2\sqrt{d}}\sum_{j\tilde{\delta}\sigma} e^{-i(\mathbf{q}/2+\mathbf{Q})\cdot\tilde{\delta}} e^{-i\mathbf{q}\cdot\mathbf{R}_j} c_{j\sigma}^\dagger c_{j+\tilde{\delta}\sigma}. \end{aligned} \tag{5}$$

The wavevector $\mathbf{Q}$ determines the symmetry of the light scattering: $\mathbf{Q} = 0$ for $A_{1g}$ scattering and $\mathbf{Q} = (\pi, 0, \pi, 0, \ldots)$ for $B_{1g}$ scattering. In Eq. (5), we used the notation $\varepsilon(\mathbf{k}) = -t^* \lim_{d\to\infty}\sum_{i=1}^d \cos\mathbf{k}_i/\sqrt{d}$ for the bandstructure, $\tilde{\delta}$ for a nearest-neighbor translation vector, and $\mathbf{R}_j$ for the position vector of the $j$th lattice site. Note that in the case of Raman scattering, with $\mathbf{q} = 0$, the scattering operator is proportional to a sum over momentum of the number operator times a function that depends on momentum and hence commutes with the kinetic energy operator; the sum rule depends solely on the multiple commutator with the potential energy operator for Raman scattering.

Evaluation of the double commutator is straightforward, but lengthy. In momentum space, we find

$$\begin{aligned}[\mathcal{O},[\mathcal{H},\mathcal{O}]] = \sum_{\mathbf{k}\sigma}\Big\{&[\varepsilon(\mathbf{k}-\mathbf{q}) - \varepsilon(\mathbf{k})]\varepsilon^2\Big(\mathbf{k} - \frac{\mathbf{q}}{2} + \mathbf{Q}\Big) \\ &+ [\varepsilon(\mathbf{k}+\mathbf{q}) - \varepsilon(\mathbf{k})]\varepsilon^2\Big(\mathbf{k} + \frac{\mathbf{q}}{2} + \mathbf{Q}\Big)\Big\} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} \\ &+ U\sum_{\mathbf{k}\mathbf{q}'\mathbf{p}\sigma} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}-\mathbf{q}'\sigma} c_{\mathbf{p}-\sigma}^\dagger c_{\mathbf{p}+\mathbf{q}'-\sigma} \\ &\times\Big\{-\varepsilon\Big(\mathbf{k} - \frac{\mathbf{q}}{2} + \mathbf{Q}\Big)\Big[\varepsilon\Big(\mathbf{k} - \frac{\mathbf{q}}{2} + \mathbf{Q}\Big) \\ &- \varepsilon\Big(\mathbf{k} - \mathbf{q}' - \frac{\mathbf{q}}{2} + \mathbf{Q}\Big) + \varepsilon\Big(\mathbf{p} + \frac{\mathbf{q}}{2} + \mathbf{Q}\Big) \\ &- \varepsilon\Big(\mathbf{p} + \mathbf{q}' - \frac{\mathbf{q}}{2} + \mathbf{Q}\Big)\Big] \\ &+ \varepsilon\Big(\mathbf{k} - \mathbf{q}' + \frac{\mathbf{q}}{2} + \mathbf{Q}\Big)\Big[\varepsilon\Big(\mathbf{k} + \frac{\mathbf{q}}{2} + \mathbf{Q}\Big) \\ &- \varepsilon\Big(\mathbf{k} - \mathbf{q}' + \frac{\mathbf{q}}{2} + \mathbf{Q}\Big) + \varepsilon\Big(\mathbf{p} + \frac{\mathbf{q}}{2} + \mathbf{Q}\Big) \\ &- \varepsilon\Big(\mathbf{p} + \mathbf{q}' - \frac{\mathbf{q}}{2} + \mathbf{Q}\Big)\Big]\Big\}. \end{aligned} \tag{6}$$

When evaluating the expectation value of the operator in Eq. (6), the first term is simple to evaluate, since it can be expressed in terms of single-particle Green's functions; the second term (proportional to $U$) is more complicated, since it involves a two-particle expectation value, and hence it requires the irreducible two-particle charge and spin vertices to solve the corresponding Bethe–Salpeter equation. While this is possible to do in principle, it is difficult to find a robust numerical technique to evaluate such a term efficiently. Note that when $\mathbf{q} = 0$, the first term vanishes, as it must, but the second term only has minor simplifications. A careful inspection of Eq. (6) also shows that there are spin flip processes present in the second piece; for example, the term with $\mathbf{p} + \mathbf{q}' = \mathbf{k}$ includes both a spin-raising and a spin-lowering operator. It does not look like such terms vanish when summed over momentum (if

$\mathbf{q} \neq 0$), so the operator should involve both the charge and the spin degrees of freedom.

In real space, the operator becomes

$$
\begin{aligned}
[\mathcal{O},[\mathcal{H},\mathcal{O}^{\dagger}]] = & -\frac{t^{*3}}{8d\sqrt{d}} \sum_{i\bar{\delta}\bar{\delta}'\bar{\delta}''\sigma} c^{\dagger}_{i\sigma} c_{i+\bar{\delta}+\bar{\delta}'+\bar{\delta}''\sigma} \\
& \times [\mathrm{e}^{\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}} (\mathrm{e}^{\mathrm{i}\mathbf{q}\cdot\bar{\delta}'} - 1) \mathrm{e}^{\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}''} + cc.] \\
& + \frac{Ut^{*2}}{4d} \sum_{i\bar{\delta}\bar{\delta}'\sigma} \{ c^{\dagger}_{i\sigma} c_{i+\bar{\delta}\sigma} \\
& \times [\mathrm{e}^{\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i+\bar{\delta}-\sigma} c_{i+\bar{\delta}-\bar{\delta}'-\sigma} \\
& - \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i-\sigma} c_{i-\bar{\delta}'-\sigma} \\
& - \mathrm{e}^{\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i+\bar{\delta}+\bar{\delta}'-\sigma} c_{i+\bar{\delta}-\sigma} \\
& + \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i+\bar{\delta}'-\sigma} c_{i-\sigma}] \\
& + [\mathrm{e}^{\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i+\bar{\delta}-\sigma} c_{i+\bar{\delta}-\sigma} \\
& - \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i-\sigma} c_{i-\sigma} \\
& - \mathrm{e}^{\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2-\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i+\bar{\delta}-\bar{\delta}'-\sigma} c_{i+\bar{\delta}-\bar{\delta}'-\sigma} \\
& + \mathrm{e}^{-\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}} \mathrm{e}^{\mathrm{i}(\mathbf{q}/2+\mathbf{Q})\cdot\bar{\delta}'} c^{\dagger}_{i-\bar{\delta}'-\sigma} c_{i-\bar{\delta}'-\sigma}] \\
& \times c^{\dagger}_{i\sigma} c_{i+\bar{\delta}-\bar{\delta}'\sigma} \}. \qquad (7)
\end{aligned}
$$

While this form appears more complex than in Eq. (6), it does show that the operator involves correlated hopping and correlated spin-flip processes, when one examines all possibilities for the nearest-neighbor translation vectors.

The inelastic light scattering sum rule depends explicitly on the model that describes the material of interest (because of the commutator with $\mathcal{H}$). In the case of the Falicov–Kimball model [7], an explicit form for the sum rule can be found in terms of the single-particle Green's functions and self-energies [1]. When this expression is evaluated, we find that the temperature dependence is weak at low temperature. This is because the expectation value depends on electronic energies, which do not vary significantly at low temperature. Similar results hold for the *f*-sum rule in an optical conductivity measurement. So we expect the temperature dependence to be weak for the Hubbard model, except possibly when one is close to the metal–insulator transition, where the system develops a renormalized low-energy scale. We have not yet been able to evaluate Eq. (6) explicitly for the Hubbard model, but we can indirectly investigate the inelastic light scattering sum rule for the Hubbard model in the $B_{1g}$ channel ($\mathbf{Q} = 0$), by integrating the Raman data calculated with the numerical renormalization group [8], to test to see whether the numerical data is consistent with the underlying sum rule. In Fig. 1, we plot these results for three different values of $U$ at half filling: (i) $U = 2.12$ which is a strongly correlated metal; (ii) $U = 3.54$ which is near the Mott transition; and (iii) $U = 4.24$ which is a small-gap strongly correlated insulator. Note how the overall sum rule tracks with $U$ due to the prefactor of $U$ since $\mathbf{q} = 0$, and how the sum rule typically flattens at low temperature, and picks up stronger temperature dependence as $T$ increases. Like in the Falicov–Kimball model, we find a $T$-dependence that decreases as $T$ increases, but here there seems to be a small region at the lowest temperatures, where the sum rule drops as $T \to 0$. We expect the numerical errors to be similar to the size of the symbols.

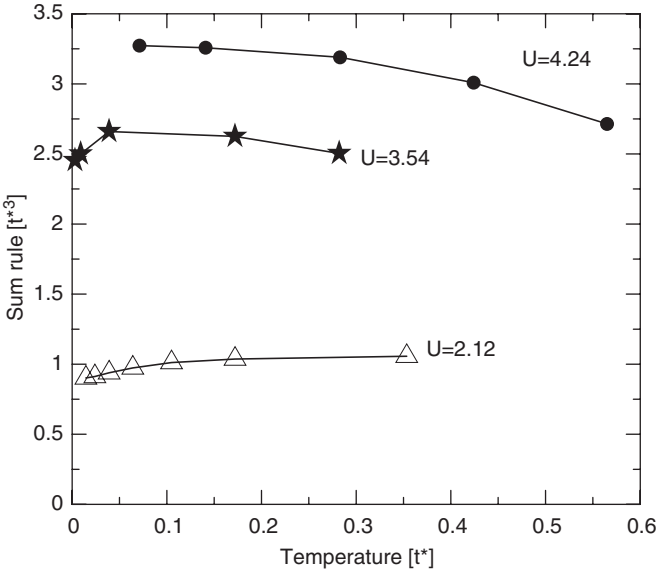


Fig. 1. Data for the sum rule in the Hubbard model at half filling. We take the Raman scattering results, multiply by the frequency, and integrate. Since the sum rule holds, the integral is equal to the operator average of Eq. (6) with $\mathbf{q} = 0$. The three curves correspond to $U = 2.12$, 3.54, and 4.24.

We plot similar results, but now in the correlated metal phase with $\rho_e = 0.9$. The differences with Fig. 1 at half filling are most significant for the larger $U$ values where the system is now metallic. Note how the sum rule drops significantly in overall magnitude when we dope off of half filling. These curves also show a slight drop at the lowest temperatures. It is interesting that the sum rule is acting as we expect, being able to give us a quantitative measure of the strength of the correlations (or the size of the potential energy effects in the system). The more correlated the system, the larger the sum rule is. Taken in this vein, the small downturn at the lowest temperatures could be arising from the development of the low-temperature coherence of a Fermi liquid which can be viewed as a reduction in the correlations. This is supported by the fact that the coherence temperature is approximately 0.1–0.15 for the largest two $U$ values and approximately 0.2 for the lowest $U$ value (as estimated from the $T$-evolution of the local DOS).

Finally, we perform the same analysis on the experimental Raman data for $SmB_6$ [1,9]. This material is an excellent candidate material because (i) there are no significant resonant scattering effects, and (ii) there is a clear separation between the low-energy, strongly correlated band, and the higher energy bands. Note how the sum rule has a generic shape that is similar to those of the

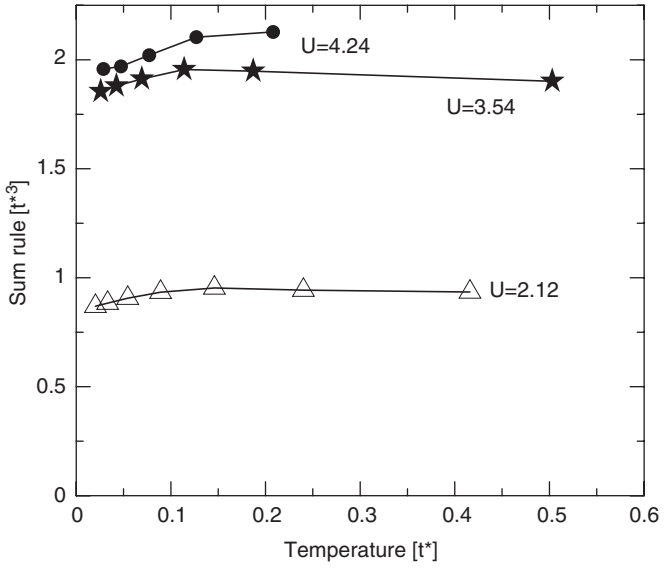


Fig. 2. Data for the sum rule in the Hubbard model at $\rho_e = 0.9$. The three curves correspond to $U = 2.12$, 3.54, and 4.24.

Hubbard model—it is relatively flat, with a slight downturn at the lowest temperatures. One interesting application of this sum rule is that it could be employed to track relative changes of the electron correlation as one varied the pressure, or the chemical doping of a material (compare Figs. 1 and 2). This may provide interesting insight into the microscopic effects of the pressure or the doping.

In conclusion, we have shown that inelastic light scattering has a first-moment sum rule that involves the potential energy of the material. We examined the results explicitly for the Hubbard model, and showed that both numerical evidence, and experimental measurements on $SmB_6$ indicate that these results can be used to quantify the level of electron correlation in a given material. Note that the sum rule holds in all dimensions, since it arises from an operator identity. The similarity between the Hubbard-model results and the experimental results indicates that the correlations are probably dominated by local correlations in $SmB_6$, as expected for a three-dimensional system (Fig. 3).

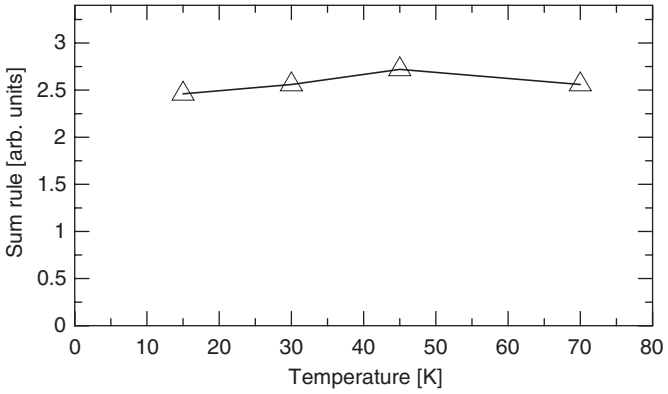


Fig. 3. Sum rule from the experimental data of $SmB_6$. Note that the results are in arbitrary units.

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