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Sherman, E.Y.; Ambrosch-Draxl, C.; Lemmens, P.; Guntherodt, G.; van Loosdrecht, P.H.M.

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Charge kinks as Raman scatterers in quarter-filled ladders

E. Ya. Sherman and C. Ambrosch-Draxl
Institute for Theoretical Physics, Karl-Franzens-University of Graz, A-8010, Graz, Austria

P. Lemmens and G. Güntherodt
2. Physikalisches Institut, RWTH Aachen, D-55099 Aachen, Germany

P. H. M. van Loosdrecht
Department of Solid State Physics & Materials Science Center, University of Groningen, 9747 AG Groningen, The Netherlands
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Charge kinks are considered as excitations in a quarter-filled charge-ordered ladder. The strength of the coupling of the kinks to the three-dimensional lattice depends on their energy. The integrated intensity of Raman scattering by the kink-antikink pairs is proportional to ϕ^5 or ϕ^4, where ϕ is the order parameter. The exponent is determined by the system parameters and the strength of the electron-phonon coupling.

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An interplay of spin, charge, and lattice degrees of freedom drives a variety of phase transitions in ladderlike compounds.[1] The best known examples of these systems are half-filled $SrCu_2O_3$ and $Sr_{14}Cu_{24}O_{41}$, where the magnetic properties are determined by $s=1/2$ spins at the Cu sites with a superexchange interaction arising from hopping via oxygen orbitals. Ca substitution on the Sr sites introduces mobile holes and leads to superconductivity in $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ under pressure.[2] The pairing interaction has a magnetic origin since it is favorable to build up a local spin singlet of two holes if the exchange interaction overcomes the kinetic energy effects.[3] Another class of ladders are the half-filled vanadates such as CaV_2O_5 and MgV_2O_5, where spins 1/2 are located at the V sites. A more interesting realization is given by NaV_2O_5 (Ref. 4) and LiV_2O_5, which have VO_5 pyramids as a common structure element with the half-filled vanadates. As follows from the chemical formula, the mean charge of V in NaV_2O_5 and LiV_2O_5 is 4.5. Although similar in stoichiometry and unit cell structure, these two compounds have strikingly different physical properties. In LiV_2O_5, the V ions are arranged in parallel chains of nonmagnetic V^{5+} and magnetic V^{4+}. In NaV_2O_5 all V ions are equivalent, making it a rare realization of the quarter-filled ladder.[4,5] Only for temperatures below $T_{co}\approx 34$ K (Refs. 6,7) the compound undergoes a charge ordering transition into a spin-gapped phase, which is due to an interplay of charge, spin, and lattice degrees of freedom.[8] It has been proposed that the spin-lattice interaction in NaV_2O_5 (Ref. 9) is due to a strong electron-phonon coupling.[10,11]

Low-energy excitations in the ladders related to charge and spin degrees of freedom and having energies less than 0.2 eV, are well suited for investigations by light scattering spectroscopy.[12,13] However, the mechanism of their Raman activity, being strongly related to the way they modulate the crystal's polarizability, requires a detailed investigation for each excitation.

Since the ladders are one-dimensional systems with strong interactions, nonlinear models appeared to be very useful for investigations of charge and lattice,[14] and magnetic[15,16] excitations. At small doping, when the interaction between the carriers is not very strong, the generalized t-J model already contains the essential physics necessary to describe the ladders.[3] In view of the large doping in the quarter filled ladders (a depletion of 1/4 hole per magnetically active site) the excitations in the charge ordered state should again be considered within a nonlinear framework. In this paper, we study the excitations in a charge ordered quarter-filled ladder system with strong coupling to the lattice. The elementary excitations are charge kinks involving ion displacements. We establish a mechanism for Raman scattering from these excitations and calculate the scattering intensity which is found to be strongly dependent on the order parameter.

In a quarter-filled ladder each rung i is occupied by a charge $Q_i=Q_i^{|L\rangle}+Q_i^{|R\rangle}=1$, where $|L\rangle$ and $|R\rangle$ denotes the left and right leg state, respectively. The electronic degree of freedom is the hopping of the charge between two ions of a rung described by a matrix element $t_\perp$. Hopping between nearest-neighbor rungs is prohibited since it requires the energy of approximately $2t_\perp$ to put electrons from two binding two-site states, with the energies of $-t_\perp$ each, to two one-site states at one rung. The wave function of an electron on a rung has the form $\psi_i=\alpha_L|L\rangle+\alpha_R|R\rangle$, where $\alpha_L^2+\alpha_R^2=1$. In the disordered phase $\alpha_L=-\alpha_R=1/\sqrt{2}$.

The total energy of the system $E_{tot}=U_{cor}+K+U_{lat}$ is the sum of the correlation energy U_{cor} due to the repulsion of electrons at nearest-neighbor sites described by V_{cor}, the kinetic energy K, and the lattice term U_{lat} which includes the electron-phonon coupling. U_{cor} is the sum over the nearest neighbor interactions of the type $V_{cor}(Q_i^{|L\rangle}Q_{i+1}^{|L\rangle}+Q_i^{|R\rangle}Q_{i+1}^{|R\rangle})$. With $\phi_i\equiv\phi_i^L=Q_i^L-1/2$, this contribution can be written as: $U_{cor}=V_{cor}\Sigma_i\phi_i\phi_{i+1}$, where $\phi_i^L=-\phi_i^R$ has been used. The kinetic energy per rung can be expressed as: $K_i=-t_\perp+2t_\perp(\phi_i^2+\phi_i^4)$. The lattice term[9,10] has the Holstein-like form $U_{lat}=C\Sigma\phi_i^l z_{i,l}+\kappa\Sigma z_{i,l}^2/2$. The elastic contribution to the energy due to a displacement $z_{i,l}$ of the ions at rung i and leg l is $1/2\Sigma_{i,l}\kappa z_{i,l}^2$, with lattice force constant $\kappa=M\Omega^2$, where M is the mass of the atom, Ω the

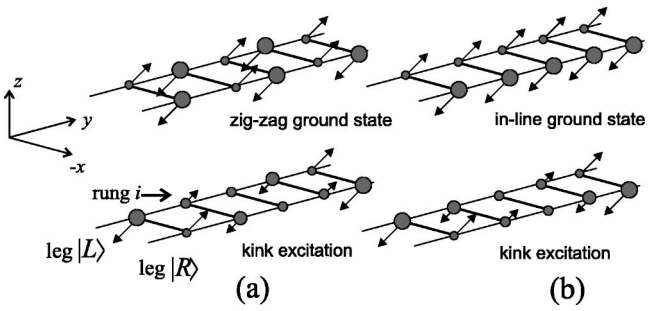

FIG. 1. (a) Displacements of V ions (proportional to the length of arrow) in the zig-zag ground state and in a kinklike excitation. The radii of the grey circles correspond to the charges of the V ions. (b) The same for the in-line ordering. (b) is presented for comparison to (a).

vibrational frequency, and C is the deformation potential. The static ion displacement minimizing $U_{\rm lat}$ is $z_i=-(C/\kappa)\phi_i$.

The equilibrium state of the system minimizes $E_{\rm tot}$. The result is a zigzag ordered state[17–19] with the order parameter $\phi_i=\phi_{\rm lat}(-1)^i$, where $\phi_{\rm lat}^2=[2(V_{\rm cor}-t_\perp)+V_{\rm lat}]/4t_\perp$, and $V_{\rm lat}=C^2/\kappa$. The subscript ''lat'' serves as a reminder that the lattice is involved in the charge ordering. The phase transition occurs when $2V_{\rm cor}+V_{\rm lat}>2t_\perp$.

In order to study the dynamics, the energy $E_{\rm tot}$ is rewritten in a continuum approximation, with a time-dependent order parameter $\phi(y,t)$ that slowly changes along the ladders. The energy acquires a form typical for the standard ϕ^4 field[20]

$$E[\phi]=V_{\rm cor}a_\parallel^2\int_{-\infty}^{\infty}[(\partial\phi/s_{\rm lat}\partial t)^2+(\partial\phi/\partial y)^2 + \lambda(\phi^2-\phi_{\rm lat}^2)^2/2]\frac{dy}{a_\parallel}, \quad (1)$$

where $1/s_{\rm lat}^2=MC^2/V_{\rm cor}a_\parallel^2\kappa^2$ with $a_\parallel$ being the lattice constant along the ladder, and $\lambda=4t_\perp/V_{\rm cor}a_\parallel^2$. The $\partial\phi/\partial t$ term in Eq. (1) arises due to the kinetic energy of the V ions which displacements follow their charges since $\partial z_i/\partial t=-(C/\kappa)\partial\phi_i/\partial t$. The ground state for $E[\phi]$ is $\phi(y)\equiv\pm\phi_{\rm lat}$. The classical excitations within this model are kinks and antikinks of the form

$$\phi(y,t)=\pm\phi_{\rm lat}\tanh\phi_{\rm lat}\sqrt{\frac{\lambda}{2}}\frac{y-ut}{\sqrt{1-u^2/s_{\rm lat}^2}}. \quad (2)$$

Here u is the kink velocity, and its energy $E_u=\gamma_{\rm lat}E_{\rm lat}$, where $E_{\rm lat}=4\sqrt{2}\sqrt{t_\perp V_{\rm cor}}\phi_{\rm lat}^3/3$, and $\gamma_{\rm lat}=1/\sqrt{1-u^2/s_{\rm lat}^2}$. The density of states of the one-kink excitation is given by: $\nu(E)=E/\pi s_{\rm lat}\sqrt{E^2-E_{\rm lat}^2}$. The displacements of ions in the ground state and in a kink excitation are shown in Fig. 1.

The above consideration is valid for a ''soft'' lattice, where ion displacements follow the electron redistribution during the kink propagation. The ''soft'' lattice condition is $\tau_{\rm k}\Omega\gg1$, where the time $\tau_{\rm k}$ characterizes the rate of the change of the order parameter. It is determined by the kink width $w_{\rm k}\sim1/\gamma_{\rm lat}\phi_{\rm lat}\sqrt{\lambda}$ as $\tau_{\rm k}\sim w_{\rm k}/u$. In the ''ultrarelativistic'' limit, where $u\to s_{\rm lat}$, the condition $\tau_{\rm k}\Omega\gg1$ cannot be fulfilled and the lattice becomes ''rigid'' due to the Lorentz contraction of the kink width $\sim1/\gamma_{\rm lat}$.

To understand the difference between a soft and a rigid lattice, let us consider the condition $\tau_{\rm k}\Omega\gg1$ in more detail. Let us assume first that the ''light'' velocity $s_{\rm lat}$ is large enough so that $\Omega/\phi_{\rm lat}\sqrt{\lambda}s_{\rm lat}\ll1$. In this case the limiting velocity $u_{\rm max}$, determined by the condition $\tau_{\rm k}\sim\Omega^{-1}$, is $u_{\rm max}\sim\Omega/\phi_{\rm lat}\sqrt{\lambda}\ll s_{\rm lat}$. At $u>u_{\rm max}$ the kinks become decoupled from the lattice and propagate on the background of ions displaced as in the equilibrium charge ordered state. The order parameter for the rigid lattice $\phi_{\rm el}^2=(V_{\rm cor}-t_\perp)/2t_\perp$ is determined by the electronic subsystem only, and the ''light'' velocity $s_{\rm el}\gg s_{\rm lat}$ since the ions are not involved in the kink motion anymore. It seems that the excitation energy drops down to approximately $E_{\rm el}=E_{\rm lat}\phi_{\rm el}^3/\phi_{\rm lat}^3$ and becomes weakly u dependent. However, due to the increase of the ''light'' velocity, the Lorentz contraction disappears and the soft lattice regime is restored. This fact implies that the lattice becomes rigid only for the fast kinks with u close to $\Omega/\phi_{\rm el}\sqrt{\lambda}$. In other words, there are no kinks in the interval $\Omega/\phi_{\rm lat}\sqrt{\lambda}\lesssim u\lesssim\Omega/\phi_{\rm el}\sqrt{\lambda}$.

In the opposite case when $\Omega/\phi_{\rm lat}\sqrt{\lambda}s_{\rm lat}\gg1$, the lattice becomes rigid when $u_{\rm max}$ is very close to $s_{\rm lat}$, such that $s_{\rm lat}-u_{\rm max}<s_{\rm lat}(\Omega/\phi_{\rm lat}\sqrt{\lambda}s_{\rm lat})^{-2}$. Therefore, the kinks involving lattice displacements are well defined up to high energies $E_{u_{\rm max}}\sim E_{\rm lat}\Omega/\phi_{\rm lat}\sqrt{\lambda}s_{\rm lat}$. Here the gap in the allowed u is determined by $s_{\rm lat}<u<\Omega/\phi_{\rm el}\sqrt{\lambda}$. If $V_{\rm cor}<t_\perp$, the kinklike electronic excitations are not well defined being strongly overdamped, and the ''rigid lattice'' electronic part of the spectrum is absent.

Now consider light scattering by the kinks. These excitations modulate the charge density and the crystal's dielectric function $\epsilon_\omega^{\beta\eta}$, where ω is the light frequency, and β and η are Cartesian indices, thereby causing inelastic light scattering at frequencies equal to the excitation energy.

The variation of the dielectric function is proportional to the square of the order parameter, and can be written for one rung as (the Cartesian indices are omitted):

$$\epsilon_\omega(\phi_i^2)-\epsilon_\omega(\phi_{\rm lat}^2)=\frac{\partial\epsilon_\omega}{\partial\phi^2}(\phi_i^2-\phi_{\rm lat}^2). \quad (3)$$

The change of the dielectric function per kink $\phi(y,t)$ from Eq. (2) is

$$\Delta\epsilon_{\rm kink}=\frac{\partial\epsilon_\omega}{\partial\phi^2}\int[\phi^2(y,t)-\phi_{\rm lat}^2]\frac{dy}{a_\parallel}=-2\sqrt{2}\frac{\partial\epsilon_\omega}{\partial\phi^2}\frac{\phi_{\rm lat}}{\gamma_{\rm lat}\sqrt{\lambda}a_\parallel}. \quad (4)$$

With increase of the energy, $\Delta\epsilon_{\rm kink}$ decreases due to Lorentz contraction of the kink width.

Because of the very small photon wave vector, Raman scattering probes excitations with zero net momentum. Therefore, the Raman active quasiparticles are kink-antikink pairs with velocities u and $-u$, respectively. The corresponding contribution of the kink-antikink pairs to the polar-

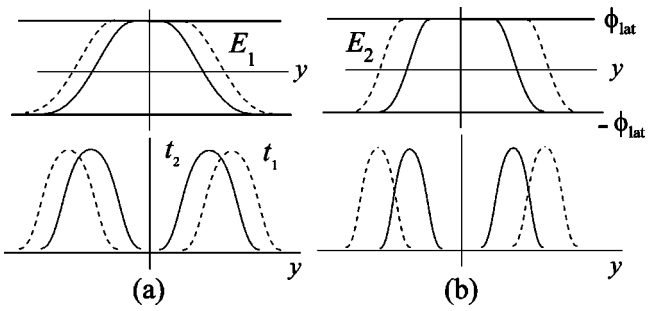

FIG. 2. The order parameter $\phi(y,t)$ in kink-antikink pairs for different kink energies (upper panel) and absolute values of the changes in the dielectric function [Eq. (3)] caused by the pairs (lower panel) for time t_2 (solid line) and $t_1 > t_2$ (dashed line). (a) Kink energy E_1, (b) kink energy $E_2 > E_1$.

izability $\epsilon_\omega^{\beta\eta}$ is shown in Fig. 2 for two different kink energies. The measured spectral density of the scattered light $\rho(E)$ as a function of energy transfer to the system (Raman shift) is proportional to the probability of the excitation of the pair with the total energy $E=2E_u$. Since the Raman scattering is a process of the decay of the incident photon into a continuum consisting of the scattered photons and electronic excitations, its probability is given by Fermi's golden rule as

$$\rho_{\rm kink}(2E) = 2\pi\nu(E)(\Delta\epsilon_{\rm kink})^2 = 16\left(\frac{\partial\epsilon_\omega}{\partial\phi^2}\right)^2 \frac{\phi_{\rm lat}^2}{\lambda a_\parallel^2 s_{\rm lat}\gamma_{\rm lat}\sqrt{\gamma_{\rm lat}^2-1}}. \quad (5)$$

The spectral density of the scattered light in Eq. (5) has a threshold at $2E_{\rm lat}$ (Ref. 21) and decreases at $E \gg E_{\rm lat}$ as E^{-2}. The integrated intensity of the Raman continuum of kinks in the ''soft'' lattice can be written as

$$I_{\rm kink} = \int_{E_{\rm lat}}^{\infty} \rho_{\rm kink}(2E)\,dE = 8\pi\left(\frac{\partial\epsilon_\omega}{\partial\phi^2}\right)^2 \frac{\phi_{\rm lat}^2}{\lambda a_\parallel^2 s_{\rm lat}} E_{\rm lat}. \quad (6)$$

The integration in Eq. (6) was extended to infinity leading to $I_{\rm kink} \sim \phi_{\rm lat}^2 E_{\rm lat} \sim \phi_{\rm lat}^5$. This can be done if $\Omega/\phi_{\rm lat}\sqrt{\lambda s_{\rm lat}} \gg 1$, which is, as we will see below, the case for NaV_2O_5. In the opposite case of $\Omega/\phi_{\rm lat}\sqrt{\lambda s_{\rm lat}} \ll 1$, the integration should be performed up to $E(\Omega/\phi_{\rm lat}\sqrt{\lambda})$, which is close to $E_{\rm lat}$ that yields $I_{\rm kink} \sim \phi_{\rm lat}^4$.

Let us now discuss a contribution of the kink-antikink excitations to the Raman spectra of NaV_2O_5, which contains a continuum[12,13] and several phonon peaks. The changes at $T < T_{\rm co}$ manifest themselves by changes in the phonon frequencies and line shapes, and by the appearance of new intense peaks. Some of these peaks are vibrational modes while others can be attributed to magnetic excitations.[12] The modification of the continuum consists of two effects: a depletion at the spectral range up to 30 meV and a moderate redistribution of the spectral weight at higher energies. This observation implies that the Raman scattering mechanism in the ordered and disordered phases must be virtually the same, despite the different character of the excitations.

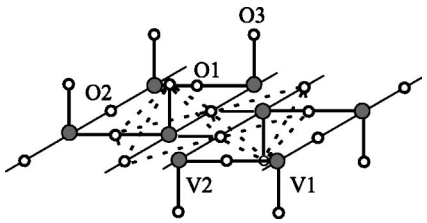

FIG. 3. Schematic plot of a part of the crystal structure of NaV_2O_5. Na ions are not shown in the figure. The Holstein-like interaction arises due to the asymmetry of the unit cell related to the position of the O3 ion.

The relevant part of the unit cell of the quarter filled ladder compound NaV_2O_5 is shown in Fig. 3. The Holstein-like electron-phonon coupling is due to the O3 ions located either above or below the V-O-V rungs[9] with the hopping matrix element $t_\perp \approx 0.35$ eV. Coupling to the lattice favors the charge ordering since $V_{\rm lat} > 0$, however it does not drive the transition itself since in the absence of correlations the lattice is stable, that, is $V_{\rm lat} < 2t_\perp$.[9]

The charge ordering parameter $\phi_{\rm lat}$ determined from the magnetic susceptibility is approximately 0.2.[22] Another possibility to find $\phi_{\rm lat}$ is provided by structural data[23] where one finds $|z_i| \approx 0.04$ Å. Assuming $C=5$ eV/Å, and $\Omega = 400$ cm^{-1} one obtains $\kappa \approx 40$ eV/Å^2, $V_{\rm lat} \approx 0.6$ eV, and $\phi_{\rm lat} = (\kappa/C)\, z_i \approx 0.3$. We accept this magnitude of $\phi_{\rm lat}$ for further estimates and for $t_\perp = 0.35$ eV, and $V_{\rm cor} = t_\perp$ obtain $E_{\rm lat} \approx 20$ meV. Therefore, one can expect a depletion in the scattering intensity for the Raman shift less than $2E_{\rm lat} = 40$ meV, in agreement with the experimental data.[12] The chosen model parameters and $a_\parallel = 4$ Å yield $\Omega/\phi_{\rm lat}\sqrt{\lambda s_{\rm lat}} \approx 10$, that is the lattice is soft up to high kink energies. This fact justifies an extension of the integration in Eq. (6) to infinity. Considering the kinks, we neglected the interaction between different ladders. As we established above, at $V_{\rm cor} < t_\perp$, the ''rigid lattice'' part of the kink spectrum is absent. We assume that this is the case in NaV_2O_5 since $V_{\rm cor} + V_{\rm lat}/2 - t_\perp$ should be considerably smaller than $t_\perp$ to ensure a small order parameter. For this reason we did not consider scattering by the high-energy kinks uncoupled to the lattice.

To have a reference point, we compare the spectral densities of the two-kink scattering and two other Raman processes relevant for this compound. The first one is the first-order phonon Raman scattering. The estimate of spectral density at the phonon peak maximum is given by $\rho_{\rm ph} \sim (\partial\epsilon_\omega/\partial\mathcal{Q})^2 (z_0/a)^2 a_\parallel/\Gamma$, where $\mathcal{Q}$ is the ratio of the ion displacement to the characteristic lattice constant a, z_0 is the zero-point vibrational amplitude for the mode, and Γ is the phonon linewidth. The other mechanism is two-magnon Raman scattering due to the frustrated superexchange interaction. Although the scattering intensity depends on many details,[24] an estimation within the Fleury-Loudon theory[25] can be done as $\rho_{\rm magn} \sim (J/E_0)^2 a_\parallel/J$, where J is the superexchange and E_0 is of the order of magnitude of the interband transition energy. By taking into account that $s_{\rm lat} a_\parallel^{-1} \sim \Omega$, the ratio of intensities can be estimated as

$$\rho_{\rm kink}:\rho_{\rm ph}:\rho_{\rm magn}\sim\left(\frac{\partial\epsilon_\omega}{\partial\phi^2}\right)^2\frac{\phi_{\rm lat}^2}{\Omega}:\left(\frac{\partial\epsilon_\omega}{\partial\mathcal{Q}}\right)^2\left(\frac{z_0}{a}\right)^2\frac{1}{\Gamma}:\left(\frac{J}{E_0}\right)^2\frac{1}{J}. \tag{7}$$

An reasonable value for $\partial\epsilon_\omega/\partial\phi^2$ is about 0.1, that is for the fully ordered state the change in the dielectric function would be about 0.1. This is consistent with the ellipsometric data on NaV_2O_5.[26,27] Equation (7) does not contain an evident small parameter which could allow to discriminate reliably intensities of the different contributions. Dominating phonon peaks arise due to their small damping $\Gamma\ll\Omega\sim J$ rather than due to high efficiency of the phonon Raman scattering. For realistic parameters $\partial\epsilon_\omega/\partial\mathcal{Q}=1$, $(z_0/a)^2=10^{-4}$, and $\Gamma=2$ meV, $\rho_{\rm ph}$ at the phonon peak is of the same order of magnitude as $\rho_{\rm kink}$ at $E\sim 2E_{\rm lat}$. This corresponds well to the experimental data,[12] especially for the light polarized along the rungs, assuming that the continuum in NaV_2O_5 is formed by the kink-antikink excitations. The two-magnon mechanism is responsible for the Raman background in LiV_2O_5, where the kink-antikink pairs cannot be excited due to long-range order of nonequivalent V^{4+} and V^{5+} ions. In this compound the continuum is weak compared to phonons,[28] however, their relative spectral densities are of the same order of magnitude as in NaV_2O_5 when the incident and scattered light are polarized along the ladders.

At $T>T_{\rm co}$ the low-energy electronic excitations are long-ranged overdamped fluctuations of the order parameter coupled to the dynamical lattice distortion. These excitations have the same mechanism of Raman scattering as the kink-antikink pairs considered above, that is modulation of the polarizability due to intrarung charge fluctuations leading to a broad intense continuum. The fluctuations of the order parameter in the form of dynamical charge ordering persist up to $T\sim 80$ K, as it was observed in Raman spectroscopy,[12] structural x-ray scattering,[29] and spin resonance[30] experiments. However, at present, it is hard to separate the contributions of the charge fluctuations and the two-magnon scattering to the Raman continuum.

In conclusion, we investigated the low-energy kink dynamics and Raman scattering by kink-antikink excitations in the charge ordered phase of a quarter-filled ladder with strong coupling to the lattice and considered NaV_2O_5 as an example. The spectral density of the scattered light and the overall scattering intensity by kink-antikink pairs very strongly depends on the charge order parameter. The mechanism corresponds well to three observed features of the continuum: (i) spectral range, (ii) depletion at Raman shifts less then 30 meV, and (iii) relative intensity to phonons.

Note added in proof. Recently, R. Valenti *et al.*[31] reported a result of calculation of the electronic structure of LiV_2O_5 done within the density functional theory. The results have shown that a quarter-filled ladder with the in-line charge ordering parameter $\phi_{\rm lat}\approx 0.2$ can be a relevant model for this compound. The contribution of the excitations corresponding to the fluctuations of the charge ordering in LiV_2O_5 to the Raman spectra is an interesting problem that remains to be investigated.

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