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On the Theory of Two-Magnon Raman Scattering

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We propose a theory of two-magnon Raman scattering, taking into account magnon-phonon interactions. The theory explains the anomalous shape of the line and its temperature dependence. We show that acoustic phonons are responsible for the asymmentry of the line, *i.e.* the low and high energy tails, as well as the temperature dependence. On the other hand, the width of the peak is determined by a magnon-vibron interaction, where the vibron is created between nearest-neighbour spin-flips.

KEY WORDS: Cuprates; Raman; two-magnon scattering; magnons; phonons.

Raman scattering (2MRS) spectra [1] cannot be explained by a spin-only model [2], although a phenomenological magnon damping [3] seems to provide a fit to experiment. Based upon this and their experimental results for the temperature (T) dependence of the 2MRS line, Knoll *et al.* [4] proposed spin-lattice interactions as the origin of the damping. A more precise calculation of this damping, but no lineshape [5], and a numerical determination of the zero temperature line due to static phonons/disorder [6] have lent credance to this suggestion. We here present a calculation of the line as a function of T using dynamical phonons [7].

We start from the spin-$\frac{1}{2}$ nearest neighbour (n.n.) Heisenberg antiferromagnet on a square lattice, with lattice constant l. The spin-lattice interaction originates from the inter-site distance dependence of the exchange coupling (J), which causes it to alter as the lattice waves distort the equilibrium ionic arrangement. Explicitly, for n.n. sites i and j, $J \to J_0 + \bar{r}_{ij} \cdot (\bar{Q}_i - \bar{Q}_j) J_1$, where J_0 is J in the undisturbed lattice, $\bar{Q}_{i,j}$ are the displacements at sites i,j and J_1 is the spin-lattice coupling constant, while $\bar{r}_{ij}$ is the unit vector between the sites.

Our aim is to produce an effective model of the Raman excitation coupled to phonons. We first express the lattice waves by phonons and the spins in terms of magnons, using the Holstein-Primakoff and Bogolioubov transformations. The Raman excitation can be viewed as a pair of n.n. flipped spins in a Néel background and has a classical energy $\varepsilon = 3J_0$, implying some binding of the spins. Short wavelength magnons are therefore dominant and we can expect Brillouin zone corner magnons to be particularly important. Such magnons have an approximately quartic dispersion and this leads to our most important approximation for we will treat the Raman excitation as an energy level at ε.

We now simplify the excitation-phonon interaction. The magnon-phonon interaction is non-diagonal in the magnons, but approximating it to be so we find that only acoustic modes couple to the excitation in the long wavelength limit. Additionally, because the excitation appears as a defect in an ordered background, it also couples to the optical mode in which n.n. spins move in opposite directions to each other. We also call this the *vibron* mode, for it can be thought of as an internal mode of the Raman excitation.

For the effective Lagrangian we have, therefore,

$$L = \bar{a}(\tau)\left[\partial_\tau + \varepsilon\right]a(\tau) + L_0\left(Q_{ac}, Q_{op}\right) + \left[\int_{q<q_D} \frac{d^2\bar{q}}{(2\pi l)^2} g_{ac} Q(\tau,\bar{q}) + g_{op} Q_{op}(\tau)\right]\bar{a}(\tau)a(\tau) \tag{1}$$

denoting the excitation, acoustic and optical phonons by $a(\bar{a})$ Q_{ac} and Q_{op}, respectively, and denoting the relevant coupling constants by g_{ac} and g_{op}; L_0 is the Lagrangian of the free phonons and $q_D = \omega_D / c$, ω_D

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being the Debye frequency, as we take the dispersion $\omega = cq$ for the acoustic mode; the frequency of the.vibron will be ω_{op}. and we have followed [8] in the manner of coupling it to the excitation.

The Raman spectrum follows from the two magnon Green function, a product of two single-particle functions in the zeroth approximation, given by the single particle function of the Raman excitation, $G(\tau)$. To find $G(\tau)$, we exploit the fact that the temperature range of interest satisfies $T << \varepsilon$. This allows us to approximate $G(\tau)$ by applying a method developed by Schwinger [9,7], as by doing so we can integrate over the excitation and phonons exactly. The propagator is then found to be given by $G(\tau) = -\theta(\tau)e^{-\varepsilon\tau + q(\tau)}$,where $q(\tau) = q_{ac}(\tau) + q_{op}(\tau)$, which describes the influence of the phonons on the excitation.

We take the 2MRS line to be proportional to the spectral density, ρ, of the excitation. The spectral density is given by

$$\rho(\omega) = 2\tanh\left(\frac{\beta\omega}{2}\right)\mathrm{Re}\left[\int_0^\infty e^{i(\omega-\varepsilon)t + q(it) - \gamma t} dt\right], \quad (2)$$

where we have introduced the damping γ. We take $\gamma = 0.14 J_0$, as this corresponds to the width of the $T = 0$ line in [2] due to spin-spin interactions only.

The acoustic phonon-excitation subsystem is specified by ω_D and a new temperature scale $T_0 \propto J_1^{-2}$. From [8], we parameterize the vibron-excitation subsystem by ω_{op} and Δ_{op}, the 'polaron' shift of the energy level. Individually the phonon modes cannot reproduce the 2MRS line: the acoustic mode is incapable of producing a broad line and the vibron of producing one asymmetric. Combined, we can obtain all the qualitative features of the 2MRS line: (i) a width $\sim 1000 cm^{-1}$ at low temperatures and $\sim 2000 cm^{-1}$ at $600K$ (ii) asymmetry, (iii) reduced dependence of the width on J_0, (iv) a T-dependent peak intensity and (v) high energy tails which merge over a wide range of temperatures. A sample result is shown in Fig.(1), where we have also taken $\varepsilon = 3500K$.

We conclude that the interaction of phonons with the antiferromagnetic fluctuations in the CuO_2 planes explains the anomalous features of the 2MRS line. Such a conclusion can be tested by means of an isotope effect (Cu or O substitution) in 2MRS. It should also be possible to use the pressure dependence of 2MRS as a proof of the theory.

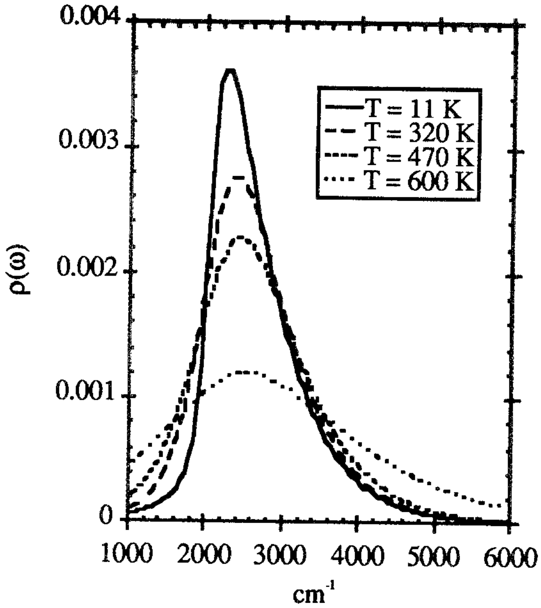

Fig. 1. The 2MRS line/$\rho(\omega)$ at the same temperatures as in [4]. We have taken $T_0 = 150K$ and $\omega_D = 450K$, along with $\omega_{op} = \Delta_{op} = 200K$.

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