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Signatures of nonlinear magnetoelectricity in the second harmonic spectra of SU(2) symmetry-broken quantum many-body systems

ABHIROOP LAHIRI* and SWAPAN K PATI

Theoretical Sciences Unit, Jawaharlal Nehru Centre for Advanced Scientific Research, Jakkur 560 064, India
*Corresponding author. E-mail: lahiri.abhiroop@gmail.com

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Abstract. Quantum mechanical perturbative expressions for second-order dynamical magnetoelectric (ME) susceptibilities have been derived and calculated for a small molecular system using the Hubbard Hamiltonian with SU(2) symmetry breaking in the form of spin–orbit coupling (SOC) or spin–phonon coupling. These susceptibilities will have signatures in second harmonic generation spectra. We show that SU(2) symmetry breaking is the key to generate these susceptibilities. We have calculated these ME coefficients using the full many-body basis and solved the Hamiltonian matrix for low-lying excited states using Lanczos method. Varying the Hubbard term along with SOC strength, we find spin and charge and both spin–charge dominated spectra of dynamical ME coefficients. We have shown that intensities of the peaks in the spectra are the highest when the magnitudes of Hubbard and SOC strengths are in a similar range.

Keywords. Nonlinear magnetoelectricity; SU(2) symmetry; Hubbard model; perturbation theory.

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1. Introduction

Nonlinear optical processes had been a topic of great interest in the previous decades due to the emergence of various intriguing physical phenomena because they have extensive applicability in various fields such as optical communications [1–3], optical sensors [4] and also for understanding different properties of materials. Optical phenomena such as higher harmonic generation have been used to get information about the crystallographic structure of materials [5]. There have been several experiments to show that the passage of light through a nonlinear medium gives finite higher-order polarisations involving higher powers of electric field. Such nonlinear susceptibilities have been measured experimentally and also calculated theoretically in crystalline materials as well as organic molecules [6]. But little is known about the mechanism of the magnetic field working in unison which may give rise to magnetoelectric susceptibilities. There have been experiments as well as classical theoretical studies showing optically induced magnetism in dielectric media [7–9]. Recent experiments [10–12] show that the magnetic field of light, despite being very small, can affect the second harmonic generation spectra of some molecules, thus showing magnetoelectric behaviour. A quantum theoretical insight has been given by Rand [13] using the density matrix approach to derive expressions for magnetically-induced polarisation. We believe that, to properly describe the effects of optically induced magnetic response in conjugated molecular systems, a many-body basis is required, since electron correlations come into play there. Although, it has been shown earlier that electron delocalisation increases harmonic yield, the Coulomb interactions can alter the charge distribution and thus can give rise to a persistent polarisation.

In this article, we have calculated the second-order nonlinear magnetoelectric (ME) susceptibilities in small molecular systems using weak incident light as perturbation using a local many-body Hamiltonian. We have performed exact diagonalisation using full many-body basis taking correlations into account. These susceptibilities will appear as small peaks in the second harmonic generation spectra of the materials. We further showed that those peak positions change with the on-site Coulomb repulsion (Hubbard U) and the spin–orbit coupling (SOC) strength. We believe that any organic molecule containing a metal with SOC showing nonlinear optical response can also show weak ME response. There have been several experimental and the-

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oretical studies on the nonlinear optical responses of organic molecules containing metals. We believe that pi-conjugated phenyl polymers containing platinum (Pt) [14] where there is prominent SOC and also metal to ligand charge transfer (MLCT) can be good candidates for exhibiting ME susceptibilites. Metalloporphyrins with nonlinear optical properties [15] can also show such responses. Organic molecular solids such as BEDT-TTF [16] having SOC as well as spin–phonon coupling [17] are capable of showing dynamical ME response.

2. Nonlinear magnetoelectric susceptibilities from perturbation theory

Following the phenomenological theory of Landau *et al* [18], the second-order ME contribution to the polarisation (P) and magnetisation (M) along the direction α, in the presence of electromagnetic field (E,B), can be written as

$$\tilde{P}_\alpha(\mathbf{E},\mathbf{B}) = -\frac{\partial \mathbf{F}}{\partial \mathbf{E}_\alpha}\bigg|_{\mathbf{B}} = \tilde{\chi}^{(2)}_{eem}\mathbf{EB} \tag{1}$$

and

$$\tilde{M}_\alpha(\mathbf{E},\mathbf{B}) = -\frac{\partial \mathbf{F}}{\partial \mathbf{B}_\alpha}\bigg|_{\mathbf{E}} = \tilde{\chi}^{(2)}_{emm}\mathbf{EB}, \tag{2}$$

where F is the free energy of the system. $\tilde{\chi}^{(2)}_{eem}$ and $\tilde{\chi}^{(2)}_{emm}$ are two types of second-order ME susceptibilities when the free energy F is proportional to **EEB** and **EBB**. In this article, we have focussed on $\tilde{\chi}^{(2)}_{eem}$, although the inferences of the results will be valid for $\tilde{\chi}^{(2)}_{emm}$ also. Light has been used as a 'probe' to find the ME coefficients. The electric and magnetic fields of light couple with the electric dipole moment and the spin, thus manifesting the non-linear ME effects in the polarisation or magnetisation. To derive the expressions for ME susceptibilities, we consider a general Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_1, \tag{3}$$

where H_0 is any arbitrary Hamiltonian and H_1 is the perturbation. H_1 can be written as

$$\begin{aligned} H_1 &= -\sum_i n_i \mathbf{E}\cdot\hat{\mathbf{r}}_\mathbf{i} - \sum_i \mathbf{B}\cdot\hat{\mathbf{S}}_\mathbf{i} \\ &= -\boldsymbol{\mu}\cdot\mathbf{E} - \boldsymbol{\nu}\cdot\mathbf{B}, \end{aligned} \tag{4}$$

where

$$\mu = \sum_i n_i \hat{\mathbf{r}}_\mathbf{i} \quad \text{and} \quad \boldsymbol{\nu} = \sum_\mathbf{i} \hat{\mathbf{S}}_\mathbf{i}.$$

Here $\mathbf{E} = \mathbf{E_0}e^{-i\omega t}$ and $\boldsymbol{B} = \boldsymbol{B_0}e^{-i\omega t}$ are the electric and magnetic fields of the incident light of frequency ω.

Using perturbation theory, we calculate the nonlinear optical coefficients following Orr and Ward [19]. The second-order correction to the polarisation is given by [20]

$$a_n^{(2)}(t) = \frac{1}{\hbar^2}\sum_m \frac{\left(\hat{\boldsymbol{\mu}}_\mathbf{nm}\cdot\mathbf{E}(\omega) + \hat{\boldsymbol{\nu}}_\mathbf{nm}\cdot\mathbf{B}(\omega)\right)\left(\hat{\boldsymbol{\mu}}_\mathbf{mg}\cdot\mathbf{E}(\omega) + \hat{\boldsymbol{\nu}}_\mathbf{mg}\cdot\mathbf{B}(\omega)\right)}{(\omega_{ng} - 2\omega)(\omega_{mg} - \omega)} \times e^{i(\omega_{mg}-2\omega)t} \tag{5}$$

where $\boldsymbol{\mu}_\mathbf{ml} = \langle\phi_m|\hat{\boldsymbol{\mu}}|\phi_l\rangle$ is the electric dipole moment and $\boldsymbol{\nu}_\mathbf{ml} = \langle\phi_m|\hat{\mathbf{S}}|\phi_l\rangle$ is the magnetic transition dipole moment. $\hat{\mathbf{S}} = S_x\hat{\mathbf{x}} + S_y\hat{\mathbf{y}} + S_z\hat{\mathbf{z}}$, S_is are spin matrices.

Clearly, the numerator of eq. (5) contains terms of the form $(\boldsymbol{\mu}\cdot\mathbf{E})(\boldsymbol{\nu}\cdot\mathbf{B})$ which leads to the coupling of the electric and magnetic fields. We calculate second-order correction to the polarisation due to the electric field and magnetic field by obtaining the expectation values,

$$\tilde{\mathbf{P}} = \frac{1}{N}\langle\psi^{(2)}|\hat{\boldsymbol{\mu}}|\psi^{(2)}\rangle, \quad \tilde{\mathbf{M}}\mathbf{s} = \frac{1}{N}\langle\psi^{(2)}|\hat{\boldsymbol{\nu}}|\psi^{(2)}\rangle\,. \tag{6}$$

Here $\psi^{(2)}$ is the second-order corrected wave function. From eqs (1) and (5) we get the second-order ME coefficients as follows:

$$\begin{aligned} &\chi^{(2)}_{ijk}(2\omega,\omega) \\ &= \frac{N}{\hbar^2}\sum_{mn}\frac{\mu^i_{gn}\mu^j_{nm}\nu^k_{mg}}{(\omega_{ng} - 2\omega + i\eta)(\omega_{mg} - \omega + i\eta)} \\ &\quad + \frac{\mu^j_{gn}\mu^i_{nm}\nu^k_{mg}}{(\omega^*_{ng} + \omega)(\omega_{mg} - \omega + i\eta)} \\ &\quad + \frac{\mu^j_{gn}\mu^k_{nm}\nu^i_{mg}}{(\omega^*_{ng} + \omega)(\omega^*_{mg} + 2\omega)}. \end{aligned} \tag{7}$$

To avoid the value of $\chi^{(2)}_{ijk}$ shooting to very large values near the poles, we have added the term $i\eta$ in the denominator. In general, when there are two different input frequencies ω_1 and ω_2, there is an intrinsic permutation symmetry $\mathcal{P}_1$ and $\chi^{(2)}_{ijk}$ should be averaged over all such permutations. This expression is similar to that obtained in ref. [18] with the numerator consisting of the product of both electric and magnetic transition dipole moments.

$$\mu^i_{gn}\mu^j_{nm}\nu^k_{mg} \equiv \langle g|\hat{\mu}^i|n\rangle\langle n|\hat{\mu}^j|m\rangle\langle m|\hat{\nu}^k|g\rangle,$$

where $|g\rangle$ is the ground state and $|n\rangle$ and $|n\rangle$ are two different excited states of the system. In the absence of SOC, the Hamiltonian commutes with $\hat{S}_z$. So two states connected by the electric dipole moment operator $\hat{\mu}_e$ (i.e. having different parity) cannot be connected by the magnetic dipole operator $\hat{\mu}_b$. Hence, the second-order ME coefficient, $\chi^{(2)}$, would be zero. Only when spin SU(2) symmetry is broken, all the states $|g\rangle$, $|m\rangle$

and $|n\rangle$ are no longer eigenstates of $\hat{S}_z$. So, coupling of the transition dipole moments could lead to a non-zero $\chi^{(2)}$. Note that in the expression for $\tilde{P}^{(2)}$, there are contributions from nonlinear optical susceptibilities $\tilde{\chi}^{(2)}_{eee}$ arising only due to the electric field of light which has very large values compared to the cross terms, $\tilde{\chi}^{(2)}_{eem}$ or $\tilde{\chi}^{(2)}_{emm}$.

In the presence of SOC, the Zeeman perturbation term in eq. (4) would be

$$\sum_i \mathbf{B}\cdot(\hat{\mathbf{L}}_\mathbf{i}+2\hat{\mathbf{S}}_\mathbf{i}) = \sum_i \mathbf{B}\cdot(\hat{\mathbf{J}}_\mathbf{i}+\hat{\mathbf{S}}_\mathbf{i}) \tag{8}$$

rather than $\sum_i \mathbf{B}\cdot\hat{\mathbf{S}}_\mathbf{i}$. Here $\hat{\mathbf{J}}$ is the total angular momentum quantum number. But, since it is a good quantum number in this process, the contribution to $\chi^{(2)}$ due to $\hat{\mathbf{J}}$ would be zero, and thus effectively the perturbation term is only due to $\hat{\mathbf{S}}$. The term could also be written as $\sum_i \mathbf{B}\cdot(2\hat{\mathbf{J}}_\mathbf{i}-\hat{\mathbf{L}}_\mathbf{i})$ and the contribution would be due to $\sum_i \mathbf{B}\cdot\hat{\mathbf{L}}_\mathbf{i}$. But the results would not change. Since $\hat{\mathbf{L}}_\mathbf{i}$ and $\hat{\mathbf{S}}_\mathbf{i}$ are coupled, taking the one with the smallest number of eigenstates would suffice.

3. The model

We consider a one-dimensional (zig-zag) chain having alternate sites with SOC (figure 1). The zig-zag nature of the system ensures that it can be exposed to an extra dimension of electric field. For simplicity, we have considered only the z-component, L_z, of the orbital angular momentum. $|L=\pm\frac{1}{2}\rangle$ are the two eigenstates of the z-component of the orbital angular momentum operator $\hat{L}_z$ with quantum number $l=\frac{1}{2}$. The single orbital Hubbard model for fermions has 4 degrees of freedom per site, which can be represented by $|c\rangle$. Hence, a fermionic site with SOC will have eight possibilities, $|c\rangle\bigotimes|L=\pm\frac{1}{2}\rangle$. We have considered the unperturbed Hamiltonian as

$$\hat{H}_0 = \sum_{\langle ij\rangle,\alpha,\sigma} t_{ij} d^\dagger_{i\alpha\sigma} d_{j\alpha\sigma} + \text{h.c.} + U\sum_{i,\alpha} n_{i\alpha\uparrow}n_{i\alpha\downarrow} + \lambda\sum_i \vec{L}_i\cdot\vec{S}_i, \tag{9}$$

where $d_{i\alpha\sigma}=(c_{i\sigma}\bigotimes\hat{L}^z)_{\alpha\alpha}$. α is the pseudospin index for two eigenstates of L_z ($+1/2$ and $-1/2$) and σ is the index for the spin. Here, $n_{i\alpha}=c^\dagger_{i\alpha}c_{i\alpha}$ is the number operator. t_{ij} and U are respectively the hopping and Hubbard parameters. λ is the strength of SOC. The term $\sum_i \mathbf{L_i}\cdot\mathbf{S_i}=\sum_i L^z_i S^z_i + L^+_i S^-_i + L^-_i S^+_i$ is considered explicitly to break the SU(2) symmetry. The last term of the Hamiltonian can also be used for spin–hardcore boson coupling after the transformation

$$\begin{aligned} L^+ &= \hat{b}^\dagger,\\ L^- &= \hat{b},\\ L^z &= \hat{b}^\dagger\hat{b}-\frac{1}{2}, \end{aligned} \tag{10}$$

where $\hat{b}^\dagger$ and $\hat{b}$ are the bosonic creation and annihilation operators and $[\hat{b}^\dagger,\hat{b}]=1$. Here we have assumed that there is a hardcore boson on each site (either 1 boson or 0 boson). These bosons can be coupled with spins which break the SU(2) symmetry.

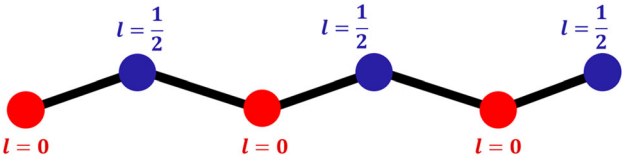

Figure 1. Schematic structure of the model system having alternating sites A and B.

4. Results and discussions

Using the above-mentioned basis set, we have diagonalised the Hamiltonian matrix exactly for a chain of six sites. We have obtained 200 lowest eigenstates using Lanczos method [21]. Using these states, ME susceptibilities are computed as given in eq. (6).

The tumbling average of these susceptibilities is computed from the expression [22,23]

$$||\chi^{(2)}|| = \frac{1}{3}\sqrt{\sum_i\sum_j |\chi^{(2)}_{ijj}+\chi^{(2)}_{jji}+\chi^{(2)}_{jij}|^2}. \tag{11}$$

These are experimentally important quantities and also define a scalar quantity for better analysis. In the Hamiltonian we have two parameters: on-site correlation U/t and SOC strength λ/t. Now, without these two terms, the solution is a plane wave with delocalised eigenstates. When $U=0$, the solution is still a charge-delocalised state. On the other hand, when $\lambda=0$ with finite U, the solution is a localised state. Thus we ask, how would the coefficients vary in the regimes (i) $U\to 0$, (ii) $U>\lambda$, (iii) $U\simeq\lambda$ and (iv) $U<\lambda$.

For a fixed value of $U/t=4.0$, we have shown in figure 2, the variation of $\chi^{(2)}_{eem}$ vs. ω (in units of $\hbar$ eV) for three different values of SOC strength λ/t. The plots are obtained by varying ω in steps of 0.01 up to 6.0. Very large value of λ is unphysical for real materials but physically realisable in systems of ultracold atoms in optical lattices where the parameters can be tuned experimentally. When $\lambda/t<U/t$, the low energy physics is

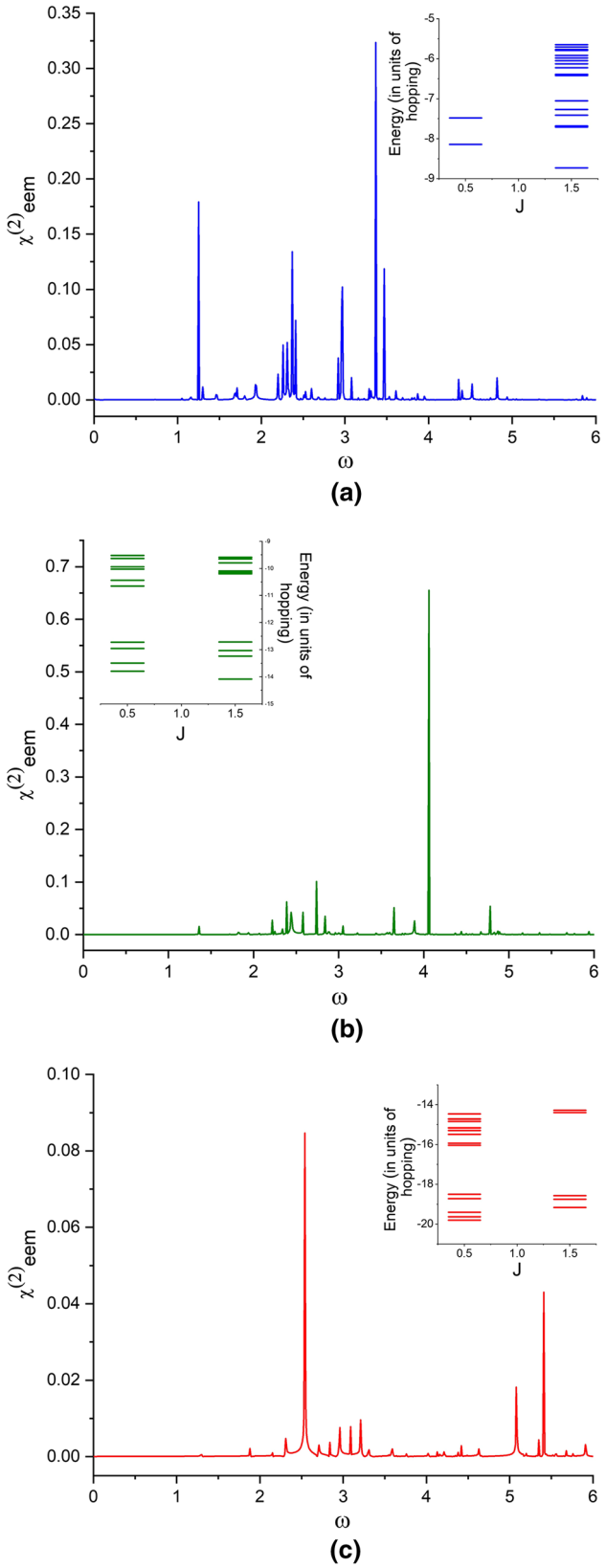

Figure 2. Plots of $\chi^{(2)}_{eem}$ vs. ω (in units of $\hbar$ eV) for $U/t = 4.0$ and $\lambda/t = 2.0, 4.0$ and 6.0. The insets show plots of energy in eV vs. total angular momentum quantum number J in each case. For $\lambda/t = 4.0$, there are equal number of states for both $J = 0.5$ and $J = 1.5$ and thus there are more degeneracies.

governed by the strength of λ. The lowest excitations are those which involve transition from $l_z = 1/2$ to $l_z = -1/2$ or vice versa. The excitations corresponding to the exchange involving Hubbard U will have higher energy. The opposite is the case in the $\lambda/t > U/t$ regime, in which the energy is lower for excitations due to magnetic exchange than those due to change in orbital angular momentum. For $\lambda/t \simeq U/t$, the energies corresponding to both the excitations are similar, that is, the excited states are highly degenerate. This is obvious from figure 3, in which figures 2a–2c are superposed. There are some less or moderately intense peaks for $\lambda/t < U/t$ and $\lambda/t > U/t$ due to less degeneracy of the excited states, and for $\lambda/t \simeq U/t$, there are less number of peaks, but a single peak with high amplitude corresponding to high degeneracy of the excited states.

The insets of figure 2a–2c also verify this result. Here we have plotted the energies of 20 lowest excited states as a function of total angular momentum J, computed from the expectation value of $\hat{J}^2$ operator ($\langle \hat{J}^2 \rangle = \langle (\hat{L} + \hat{S})^2 \rangle$). It is evident from these insets that, for $\lambda/t < U/t$, the low-energy excited states will have higher J values as the electrons prefer filling different angular momentum states rather than pairing up in one orbital. Hence, the density of states is high at $J = 1.5$. For $\lambda/t > U/t$ the exchange is preferred. Hence, the states with lower value of J, that is, $J = 0.5$ have more population of states. When $\lambda/t = U/t$, the excited states comprise excitations due to both exchange and SOC and hence

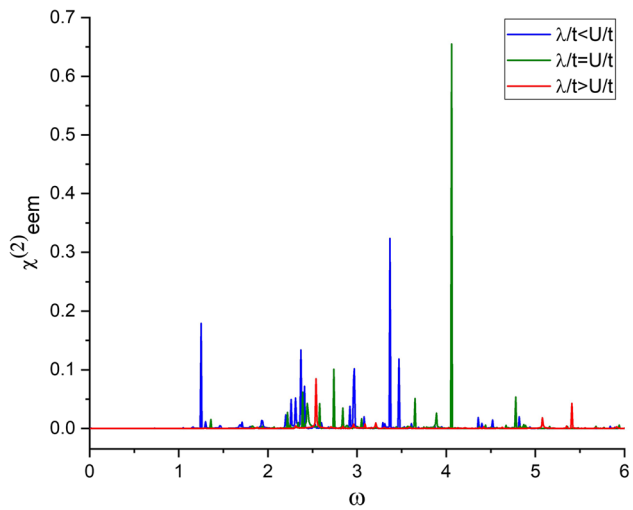

Figure 3. Combined plot of $\chi^{(2)}_{eem}$ vs. ω (in units of $\hbar$ eV) for $U/t = 4.0$ and $\lambda/t = 2.0, 4.0$ and 6.0. For $\lambda = 2.0$ and 4.0, there are more number of less intense peaks. For $\lambda/t = U/t = 4.0$, there is a single peak with high amplitude due to the high degeneracy of the excited states involving excitation corresponding to both exchange and orbital transitions.

there are almost equal number of states having $J = 0.5$ and $J = 1.5$.

The plots of $\chi^{(2)}_{eem}$ vs. ω for five different values of U/t for a fixed $\lambda/t = 0.8$, are shown in figure 4. The spin excitation cost energy $\approx \lambda$ and at most 3λ since there are three sites with SOC in our system. So we find a high-amplitude peak in the regime $U/t = 0.8$–2.4 as excitations due to U and λ have similar energies, leading to more degeneracies. For $U < \lambda$ and $U > 3\lambda$ the degeneracies are broken and so the amplitudes of the peaks decrease. Also, for higher U values, very few peaks are visible only at lower values of ω, namely, those connecting states having similar $\langle U \rangle$. This is because the spin excitations together cannot match the excitations costing energy U and so peaks at higher ω values are not probable.

From figure 4, it is also evident that there are non-zero ME susceptibilities at $U = 0$. In this regime, the model is effectively tight-binding with SOC. Though the electronic spins are completely delocalised in this case, there is no role of kinetic exchange. But the broken SU(2) symmetry in the presence of SOC leads to spin–orbital excitations between different eigenstates and hence it is sufficient to give non-zero $\chi^{(2)}_{eem}(\omega)$.

The effect of SU(2) symmetry breaking upon spin–phonon coupling [24–26] will be similar when we consider hardcore bosons, owing to the equivalence of the two, as shown in eq. (10). But in reality, there are many bosonic modes. So, one has to use Holstein–Primakoff transformations [27] to obtain $\hat{\mathbf{L}}$ operator from the bosonic operators and thus the Hamiltonian for SOC can also be used. In that case, there will be many eigenstates for $\hat{L}_z$. So, for low values of λ, there will be many possibilities for transitions among l values giving rise to many more peaks.

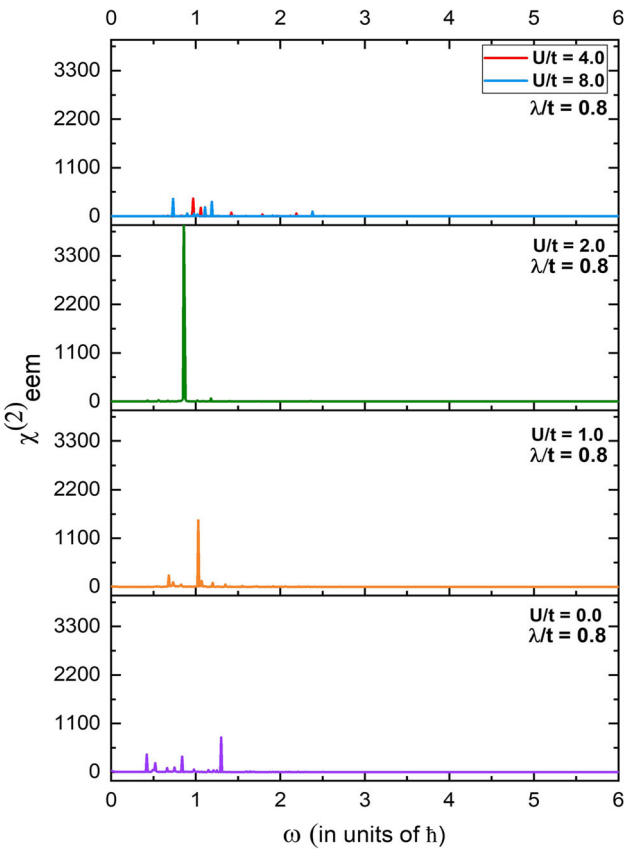

Figure 4. Plots of $\chi^{(2)}_{eem}$ vs. ω for different values of $U/t = 0.0, 1.0, 2.0, 4.0, 8.0$. For $U = 1.0$ and 2.0, the peaks have greater amplitude owing to similar excitation energies due to SOC and exchange.

5. Conclusion

In conclusion, we have shown that there can be non-zero second-order dynamical ME susceptibility $\chi^{(2)}_{eem}(\omega)$ at certain resonant frequencies in a system when the spin SU(2) symmetry is broken by spin–orbit or spin–phonon coupling. These resonant frequencies correspond to different spin and charge excitations in the case of SOC. For spin–phonon coupling, these correspond to charge excitations as well as excitations between different phonon modes. The amplitude of the peaks are very high when both the excitations are in similar energy range.

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