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Landau damping and lifting of vibrational degeneracy in metallic potassium fulleride

J. Winter and H. Kuzmany
Universität Wien, Institut für Festkörperphysik, Strudlhofgasse 4, A-1090 Wien, Austria

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Raman spectra are presented for single crystal metallic potassium fulleride at low temperatures. As compared to the undoped material dramatic changes in line position ($\Delta\omega \geqslant 80$ cm^{-1}) and linewidth ($\gamma \geqslant 60$ cm^{-1}), particularly for the two highest and the two lowest H_g modes, were observed. The lowest frequency H_g modes were found to be asymmetric but structured and could be fitted with five components which exhibit a line shift-linewidth relation. The experimental results are discussed in a model of single-particle excitation (Landau damping). Electron-phonon coupling constants λ_i were determined for all eight H_g modes using Allen's formula. The total for all λ_i was found to be 0.9 for a density of states at the Fermi level of 12 eV^{-1}.

I. INTRODUCTION

One of the many exciting properties of fullerene materials is the possibility to obtain a metallic state after doping with strong electron donors like alkali or earth alkali metals[1,2] with typical Fermi wave vectors k_F of the order of 10^7 cm^{-1}, corresponding to free carrier densities between 10^{21} and 10^{22} cm^{-3}. Some of these metals become superconductors with transition temperatures as high as 33 K.[3,4] At present most of the experimental results indicate that the mechanism for reaching the superconducting state is pairing mediated by phonons with weak or intermediate coupling.[5,6]

The questions to be discussed in this paper extend, however, beyond the problems related to superconductivity. They are rather concerned with the general single-particle excitation and its coupling to vibrational modes as visualized from an electron-phonon interaction induced line shift and line broadening. Such changes of the self energy of phonons are an important problem in solid systems with free carriers. They have been reported to some extent for heavily doped semiconductors like Si or Ge,[7,8] but are rather small, of the order of a few wave numbers, in these cases. Also, in the case of the classical semiconductors the change in phonon self energy is very often explained by a strain induced shifting of band minima and consequent repopulation rather than by a decay of the phonon into an electron hole pair. Free carrier induced changes of the self energy has been observed to an even lesser extent in metallic systems. The best known example is a line broadening observed by neutron scattering in Nb_3Sn in the normal state as compared to the linewidth in the superconducting state.[9] Such indirect evidence for a contribution of free carriers to the self energy was also found in oxidic superconductors.[10,11] Most recently an explicit line broadening and line shift was reported from a comparison between undoped La_2CaO_4 and the corresponding Sr doped compound.[12]

Several attempts have been made to determine the coupling constants between electrons and phonons in metallic fullerides from an analysis of the linewidths in vibronic spectra excited either by light[13] (Raman scattering) or by neutrons.[14] From the t_{1u} symmetry of the electrons in the conduction band a coupling is only possible to the total symmetric A_g modes and to the fivefold degenerate H_g modes. The coupling to the former is expected to be weak because of an efficient screening effect.[15] The coupling to the latter is expected to be quite strong since it allows a Jahn-Teller mechanism. For the low frequency H_g modes linewidths up to 35 cm^{-1} have been reported from the inelastic neutron experiments and up to 25 cm^{-1} from Raman experiments but the spectra were of low resolution, in particular in the case of neutron scattering where the limit in energy resolution is of the order of 5%. High frequency H_g modes could be identified neither by neutron nor by Raman scattering because of the low neutron flux at high energies in the former and because of the low scattering cross section combined with a very large bandwidth at least as evaluated from local density approximation (LDA) and frozen phonon calculations[16,17] in the latter case. Indirect evidence for the very large linewidths and very strong electron-phonon coupling induced shifts was obtained from vibrational structure in IR spectra.[18,19]

Asymmetric line shapes have been reported for the low frequency H_g modes from light scattering experiments and were suggested to originate from a Fano interference effect between scattering from the discrete phonon and a continuum of electronic transitions coupled by an electron-phonon interaction.[20–22] However, since these experiments were performed on polycrystalline material with rather broad lines it could not be excluded that the asymmetry resulted from an unresolved superposition of several split components.

Under the described circumstances it was highly desirable to obtain high resolution and high intensity Raman spectra from single-crystal K_3C_{60}. We present a detailed study of such spectra as excited with two different laser lines at low temperatures. Several dramatic results were obtained, particularly for the H_g modes.

(1) The Raman response for all A_g and all H_g modes could be observed which allowed us to evaluate the electron-phonon coupling constants for all directly coupling modes.

(2) The observed linewidths and line shifts are indeed much larger as compared to results reported so far for some of the modes. They reach values beyond 60 cm^{-1} and 80 cm^{-1}, respectively.

(3) The low frequency H_g modes lose all degeneracy and

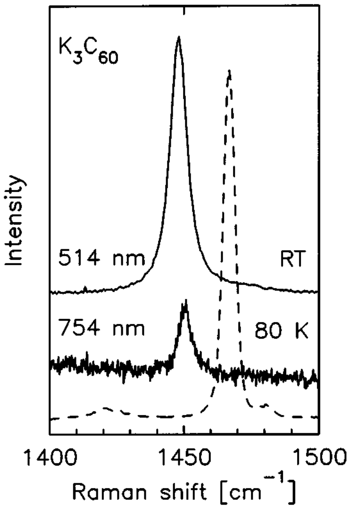

FIG. 1. $A_g(2)$ pinch mode of the K_3C_{60} phase as excited with a laser at 514 nm at room temperature and at 754 nm at 80 K, respectively. The dashed line indicates the response of an undoped crystal.

split into five components, each of which couples differently to the electrons. Linewidth and lineshift for the components of each mode are clearly related. The increase in line width by the doping process is up to a factor of 60.

(4) Raman intensities are reversed for red and green laser excitation. Strong coupling components are enhanced in the red and weak in the green and vice versa for the weak coupling components.

As reviewed above the shift and broadening of vibrational lines due to a coupling to free carriers is of great interest in various fields of solid state physics. The extremely strong effect as described here for the fullerene metal K_3C_{60} is, however, exceptional and so far unique.

II. EXPERIMENT

Single crystals were grown from highly purified C_{60} by the sublimation technique.[23] Doping was performed in a stream of hot potassium vapor, *in situ* in front of the Raman spectrometer. Two electrical contacts were applied to control the doping process *in situ* by measuring the overall resistance between the contacts. During the whole experiment the vacuum was kept to a level of better than 10^{-4} Pa.

The single-phase condition was checked by finally cooling the sample to liquid nitrogen temperature and measuring the Raman response for the pinch mode which appears at 1450 cm^{-1} for K_3C_{60}. Since this mode is known to be very sensitive to the charge on the C_{60} molecule[21] its observation and analysis can be used to check on impurity phases. Figure 1 shows the Raman response in the spectral range of the pinch mode for excitation with a green laser and with a much deeper penetrating red laser. The dashed line marks the position where the line for the undoped material is expected. From the lack of any signal at this position a phase purity of about 95% (resolution of the experiment) at least to the penetration depth of the lasers is proved.

Raman spectra were excited with an argon laser at 514.5 nm and with a TiSa laser at 754 nm with 60 W/cm^2 and 90 W/cm^2, respectively. The scattered light was analyzed and detected with a Dilor *xy* multichannel spectrometer using a spectral resolution of 3 cm^{-1}. Decomposition of the spectra into individual lines was performed with a peak-fit routine after a careful subtraction of the background originating from the laser.

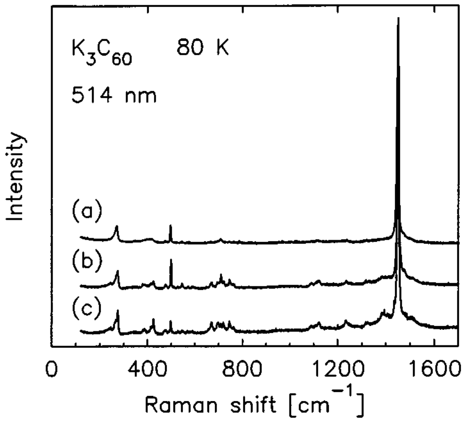

FIG. 2. Raman spectra from 100 to 1700 cm^{-1} for the K_3C_{60} phase at room temperature (a) and at 80 K (b), and (c) as excited with 514.5 nm. Spectra (b) and (c) originate from two different crystals.

III. RESULTS

Figure 2 shows Raman spectra for the single-phase crystal of K_3C_{60} as excited with the green laser at room temperature (RT) and at 80 K in the full frequency range of the first order internal vibrational modes. For the room temperature spectra the dominance of the line for the pinch mode, the lack of sizable contributions from the high frequency H_g modes, and the two low frequency features from the $A_g(1)$ and from the $H_g(1)$ mode are as known from previous experiments. There is a dramatic change in the spectra if excited at low temperature. The two spectra (b) and (c) are shown in the figure to demonstrate the amount of reproducibility of the scattering response. Several characteristic features are remarkable in the low temperature spectra. There exists a considerable oscillator strength on either side of the pinch mode which can be assigned to contributions from the high frequency H_g modes 8 and 7. At lower wave numbers two strong and partly split features appear around 1220 cm^{-1} and 1100 cm^{-1} which may be assigned to the H_g modes 6 and 5. Then, a bunch of lines appears around 700 cm^{-1} which are likely to be assigned to the H_g modes 4 and 3 and the sharp peak at 500 cm^{-1} is the well known $A_g(1)$ mode. The two features at and below 400 cm^{-1} deserve particular attention. They are characterized by a strong peak at the high frequency edge and a long tailing structure towards lower frequencies. More details about these structures can be seen in the spectra excited with the red laser as discussed below.

The low noise of the spectra allowed a detailed analysis of the various spectral ranges. Figure 3(a) gives a detailed presentation of the 1400 cm^{-1} spectral range as a whole whereas part (b) of the figure is the rest of the response after subtraction of a Voigtian profile for the contribution of the pinch mode. The full drawn lines in (b) are fits with Voigtian profiles. The broad components to the left and to the right of the central dip are considered to represent the response from the modes $H_g(7)$ and $H_g(8)$.

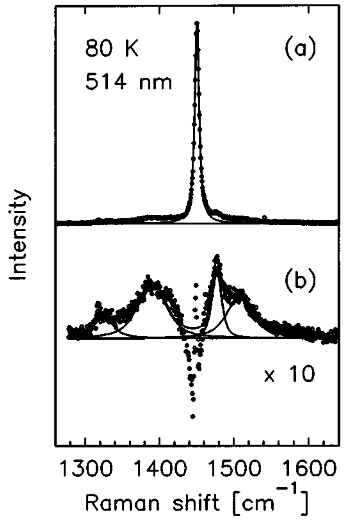

FIG. 3. Raman response of the pinch mode from single-crystal K_3C_{60} as excited at 80 K with 514.5 nm (a) and 10 times magnified signal after subtraction of the central pinch mode contribution (b). Full drawn lines are oscillator fits.

The next interesting part of the overall spectrum is the spectral range of the modes 3 and 4. It is shown in an enlarged presentation in Fig. 4 in comparison to previous results from polycrystalline material at room temperature. The low temperature spectrum consists of a large number of lines which are obviously splited components of the two H_g modes.

The two lowest frequency H_g modes are of particular interest. They are shown in Figs. 5 and 6 as excited with a green and a red laser. As can be seen in particular from the excitation with the green laser both lines again consist of a set of components with the striking character of a continuously increasing linewidth with increasing downshift of line position. The quantitative results from the line-fit procedure completely confirmed the first impression and are summarized in Table I. The table also includes similar results for the excitation with the red laser (in parentheses) as will be discussed below. The second column in the table gives center of gravity frequency for the two modes in the undoped crystal, also measured at low temperatures.

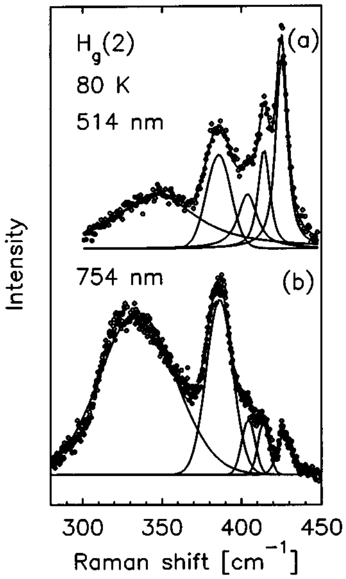

FIG. 5. Raman spectra of the $H_g(2)$ mode at 80 K as excited with a green laser (a) and with a red laser (b). The full drawn lines are computer fits which add up to the line on top of the experimental results.

At a first glance the spectra excited with the red laser [parts (b) in Figs. 5 and 6] look very different from those excited with the green laser. A detailed inspection shows that this is not so. The Raman line positions and linewidths are the same for both excitations as can be seen from the values in parentheses in Table I. The rather good agreement between the line parameters as obtained for the two lasers is evident. Only the relative intensities for the individual com

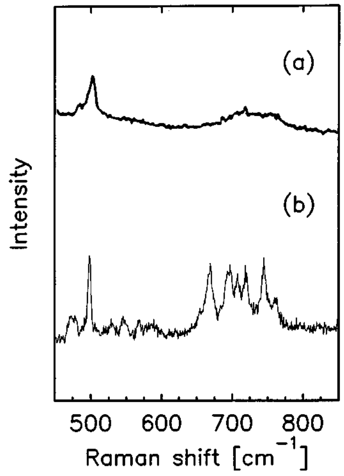

FIG. 4. Raman response in the spectral range of the H_g modes 3 and 4 from a polycrystalline material Rb_3C_{60} at room temperature (after Ref. 21) (a) in comparison to the present results from a single crystal at 80 K (b).

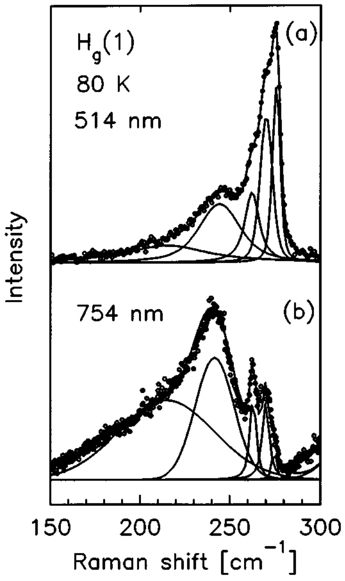

FIG. 6. Raman spectra of the $H_g(1)$ mode at 80 K as excited with a green laser (a) and with a red laser (b). The full drawn lines have the same meaning as in Fig. 5.

TABLE I. Positions and linewidths for H_g modes in K_3C_{60} as related to C_{60}. Positions and widths are as measured with 514.5 nm and 754 nm (in parentheses) excitation.

Mode	C_{60}	K_3C_{60}			
80 K	ω (cm^{-1})	ω (cm^{-1})		γ (cm^{-1})	
$H_g(1)$		275	(275)	5	(3)
		270	(270)	8	(4)
	270	262	(263)	10	(4)
		244	(242)	27	(25)
		215	(214)	54	(63)
$H_g(2)$		425	(427)	8	(10)
		414	(414)	9	(8)
	432	404	(405)	14	(10)
		386	(386)	18	(20)
		346	(335)	60	(55)

ponents are reversed. What is strong for the green laser excitation is weak for the red laser excitation.

IV. DISCUSSION

A rather strong shift of vibrational modes as a consequence of charge transfer is not unusual in molecular organic material. Such shifts have been observed in the tetrathiofulvalene (TTF) salts to reach values up to 100 cm^{-1} per charge.[24] These shifts are, however, not accompanied by a line broadening and can therefore not be correlated to free carrier effects. In contrast, for the case reported here a clear relation between line shift and line broadening is found as is required for an interaction with free carriers.

A problem for the decay of the phonons into an electron-hole pair may arise from the rather large mass of the carriers in K_3C_{60}, which is expected to be several times the free carrier mass. Considering the small q vector of the phonons involved in the light scattering process momentum conservation conditions for the decay of a phonon into an electron-hole pair may not be fulfilled for a perfectly ordered lattice.[25] However, the intrinsic merohedral disorder in the systems studied here may help to overcome this problem as was recently discussed by Schlüter *et al.*[26] Accordingly, the electron-phonon interaction may well be considered in our case as the reason for the broadening and for the shift of the lines and electron-phonon coupling constants may be deduced.

For the description of the relation between phonon linewidth and electron-phonon coupling constants for a single-particle excitation Allen's formula may be used in the form[27]

$$\gamma_i=\frac{1}{g_i}\frac{\pi}{2}N(0)\lambda_i\omega_{bi}^2 \tag{1}$$

where for each of the modes γ_i is the full width at half maximum (FWHM) of the line, $N(0)$ the density of states at the Fermi level per spin and molecule, and λ_i and g_i the dimensionless electron-phonon coupling constants and the mode degeneracy, respectively. ω_{bi} is the bare phonon frequency which means the frequency before any coupling to the electrons. The applicability of Allen's formula to metallic C_{60} was questioned several times, since Migdal's theorem concerning the ratio between bandwidth W and phonon frequency ω is not very well satisfied. However, it was shown recently by Rice and da Costa that for high enough disorder, weak coupling, and a constant density of states the relation should hold as long as $0 \leqslant \omega \leqslant W/2$.[28] In this case the total coupling which enters, e.g., the BCS relations or the MacMillan relation is given by the sum of the coupling constants for the individual modes. Thus the results given below should be valid to the extent that $\lambda\omega/W \leqslant 0.15$ is $\ll 1$.

According to the arguments given above Allen's formula will be used in the following to derive the coupling constants for the eight H_g modes. For the evaluation frequencies for the undoped system were used as the bare frequencies. This may not be a good approach for the weakly shifted modes but reasonable for shifts as high as several tens of cm^{-1}. Since the coupling constants obtained for the weakly shifted modes do not contribute significantly to the overall coupling the approach seems well justified.

Particular attention has to be paid to the two low frequency H_g modes. This part of the spectrum will be considered under the assumption that the fivefold degeneracy of the modes is lifted and each component couples with a different strength to the t_{1u} carriers. The splitting of the modes can be understood from the merohedral disorder well known for the K, Rb, and Cs derived metallic fullerides.[29] This disorder is of low enough symmetry to allow only one dimensional representations for all modes. Experimental evidence for the splitting can be deduced from the line shape pattern in the frequency range of the H_g modes 3 and 4.

Alternative explanations for the splitting are based on a Jahn-Teller type interaction. It was shown recently that this interaction can give rise to a new vibrational system with a rather large number of components, even more than five.[30] Also, a contribution to the splitting from an internal strain between the doped part of the crystal and the undoped part of the crystal cannot be excluded even though in this case a more continuous effect like a general line broadening would be expected.

Since the electron-phonon coupling constants are very sensitive to even small changes in the normal coordinates, as was shown recently from an explicit calculation by Gunnarsson *et al.*,[31] different coupling constants for the different components of the modes are not unreasonable. Further evi-

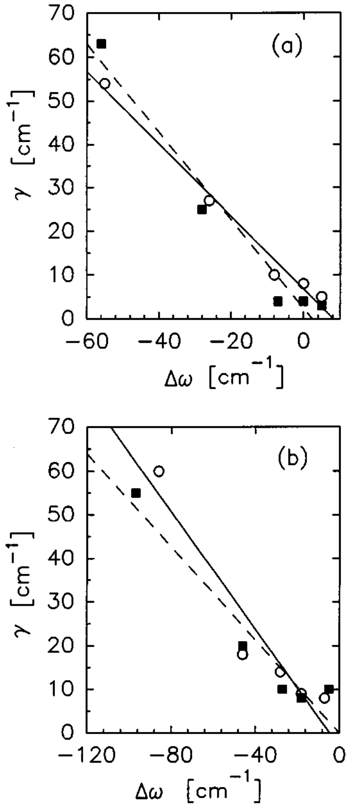

FIG. 7. Plot of linewidth γ versus observed frequency shift $\Delta\omega$ for the individual components of the mode $H_g(1)$ (a) and $H_g(2)$ (b). Open circles and full squares are for excitation with the green and the red laser, respectively.

dence for this interpretation comes from the relation between the observed linewidths and line shifts. In the framework of Allen's theory there should be a linear relation of the form

$$\gamma = -\frac{\pi}{2} N(0)\,\omega_b \Delta\omega \qquad (2)$$

between the two quantities where $\Delta\omega$ is the difference between the bare phonon frequency and the observed frequency. The experimental values as determined for the red and for the green laser excitation for the two H_g modes (Table I) are plotted in Fig. 7. Again, the result obtained for the undoped crystal is used as the bare phonon frequency. In agreement with Eq. (2) the relations between $\Delta\omega$ and γ are linear and thus support our model. The slope of the lines should yield $N(0)$. The values obtained are 8 eV^{-1} and 16 eV^{-1} for the two H_g modes, respectively. Even though these two values are quite different, both of them are within the range of reported values for $N(0)$. The discrepancy may arise from the fact that we could not use the real bare phonon frequencies for the evaluation. Geometry effects may also contribute to the shift and may be different for the two modes, thus giving a straightforward explanation for the disagreement. For the evaluation of the coupling constants as discussed below a value of 12 eV^{-1} is used for $N(0)$ in agreement with the most often reported results in the literature. Since we are finally only interested in the overall coupling constants for each mode and for all modes together the averaged linewidths for the split modes were evaluated and compiled in Table II, together with the frequencies for the pristine crystal.

The procedure for the evaluation of the linewidths was similar but more straightforward for the other H_g modes. For split modes a weighted average was used if not all five components could be determined explicitly. This is not a serious problem since the narrow lines observed for the modes $H_g(3)$ to $H_g(6)$, where not all the components could be determined, hardly contribute to the coupling. Similarly, splitting for the modes in the undoped crystal is neglected and an average value for these frequencies was assumed. The values for λ_i have been evaluated using Eq. (1). Results are shown in column 4 in Table II.

Recently coupling constants for the H_g modes have been determined from photoemission experiments on singly charged molecules in the gas phase.[31] Also, coupling constants were reported by Varma *et al.*[16] and by Schlüter *et al.*[17] from theoretical models. Sets of these results obtained for the H_g modes are included in Table II. The total coupling constant as it is, e.g., responsible for superconductivity is given by the sum of the individual coupling constants and shown in the last line of the table. According to this the results from the photoemission spectroscopy (PES) experiment appear too large which may be either due to the

TABLE II. Positions, linewidths, and electron-phonon coupling constants normalized to the density of states at the Fermi level for the eight fivefold degenerate H_g modes. The values for γ are for excitation with 514.5 nm. The columns PES, Varma, and Schlüter refer to experimental results obtained from photoemission (Ref. 31) and to calculations from Varma *et al.* (Ref. 16) and Schlüter *et al.* (Ref. 17).

Mode	ω (cm^{-1})	$\bar{\gamma}$ (cm^{-1})	$\lambda/N(E_F)$ Raman	PES	Varma	Schlüter
$H_g(1)$	270	20	0.048	0.019	0.003	0.008
$H_g(2)$	432	21	0.020	0.040	0.001	0.007
$H_g(3)$	709	8	0.002	0.013	0.001	0.004
$H_g(4)$	773	10	0.003	0.018	0.000	0.007
$H_g(5)$	1100	11	0.001	0.012	0.006	0.001
$H_g(6)$	1248	10	0.001	0.005	0.000	0.003
$H_g(7)$	1425	46	0.004	0.017	0.034	0.013
$H_g(8)$	1572	42	0.003	0.023	0.011	0.009
Σ			0.081	0.147	0.056	0.052

fact that in these experiments in effect the uncharged molecule instead of the triply chared molecule was investigated or, even more likely, due to the fact that a smaller value for $N(0)$ of only 7 eV^{-1} was used.[32] Normalizing all results from the last line in Table II to the same density of states of 12 eV^{-1} and considering a contribution of the spectral resolution of 3 cm^{-1} to the experimentally determined linewidths in the case of Raman scattering yields for the total coupling constants the values 0.075, 0.086, 0.047, and 0.061 for the four examples given in the table, respectively. Now, both experiments appear in rather good agreement and give slightly higher values for λ than the calculations.

Looking at the contributions of the individual modes to the total coupling a remarkable discrepancy remains between experimental and calculated results. Whereas from the Raman experiments the strongest contrubution comes from the two lowest H_g modes and the two highest H_g modes contribute only moderately, the calculations reveal the strongest contributions from the two highest modes and only a small contribution (Varma *et al.*) and a moderate contribution (Schlüter *et al.*) from the low frequency modes. The PES experiments distribute the contributions about equally to the two highest and to the two lowest frequencies. This discrepancy between experiment and calculation seems to remain unsolved so far. There is, however, agreement between the two different experiments and the calculations that the intermediate H_g modes do not contribute essentially to the coupling.

From the observed frequencies and the evaluated coupling constants the logarithmically averaged phonon frequency was determined as $\omega_{\log}=390\ \mathrm{cm}^{-1}$. With this value and the rather strong overall coupling obtained from the Raman results the transition temperature to superconductivity of 18 K can be evaluated from the McMillan relation

$$T_c=\frac{\omega_{\log}}{1.2k_B}\ \exp\left[-\frac{1.04(1+\lambda)}{\lambda-\mu^*-0.62\mu^*\lambda}\right] \tag{3}$$

for the screened and retarded Coulomb repulsion $\mu^*=0.18=\mu/1+\mu\ \ln(\omega_e/\omega_{\log})$. This is not an unusual value for μ^*. μ is the unretarded but screened Coulomb interaction which has the approximate value of 0.5 for a simple jellium model.[33] ω_e is a characteristic electron energy which covers the scattering range for the Coulomb interaction process. The result suggests that the high transition temperature can be obtained for characteristic electron energies of the order of 2 eV which is beyond the width of the t_{1u} band. Thus, scattering across the whole valence band is required as was discussed in Ref. 26.

The values obtained for μ^*, λ, and $\omega_{\log}$ allow us also to calculate the isotope effect as a logarithmic derivative of T_c from Eq. (3):

$$\alpha=-\frac{\partial\ \ln T_c}{\partial\ \ln M}=\frac{1}{2}\left[1-\left\{\mu^*\ \ln\frac{\omega_{\log}}{1.2k_BT_c}\right\}^2\frac{(1+0.62\lambda)}{1+\lambda}\right]. \tag{4}$$

Inserting the obtained values yields $\alpha=0.36$ in excellent agreement with the directly obtained result of 0.37 from experiments with isotope substituted C_{60}.[34]

V. CONCLUSION

Experimental results from Raman scattering on single-crystal K_3C_{60} obtained at low temperatures were used to determine the response of all eight Raman active H_g modes. The spectra revealed a strong broadening and a characteristic downshift of the modes, particularly for the two lowest frequency and for the two highest frequency vibrations. The former are split into five components with a characteristic relation between line shift and linewidth in agreement with an origin for the change of the phonon self energy by an electron-phonon interaction process.

The evaluation of the experimental data on the basis of a decay of the phonons into an electron-hole pair revealed the electron-phonon coupling constants in the framework of the applicability of Allen's theory for this interaction. The obtained coupling constants are in good agreement with recent results from photoemission experiments on singly charged molecules but slightly higher than calculated values.

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