COLLISIONAL BROADENING OF THE RB[87] HYPERFINE TRANSITION

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The width of the Rb[87] ground state hyperfine transition at 6835 MHz has been measured as a function of foreign gas density for He, Ne, Ar, N_2, and CH_4. Standard optical pumping techniques were employed. Care was taken to extrapolate the measured line widths to zero light intensity and zero microwave power. The F = 2, M_F = 0 to F = 1, M_F = 0 transition was observed in a field of a few milligauss in order to avoid broadening due to magnetic field inhomogeniety. The microwave frequency was derived from an oscillator maintained by the NBS Atomic Frequency and Time Standards Section which had a very narrow spectral width and a low drift rate.

The observed line width can be much less than the normally expected Doppler width for samples containing an alkali vapor plus a relatively inert buffer gas. This collisional-narrowing effect was originally suggested by R. H. Dicke[1] and demonstrated by Wittke and Dicke.[2] It arises from the increased time necessary for an atom to move a substantial fraction of a wavelength along the direction of propagation of the microwave field. This results in a residual Doppler width which is inversely proportional to buffer gas density.

In order to observe a high degree of collisional-narrowing, it is necessary that individual collisions be ineffective in disturbing the internal state of the atom. Collisional broadening is of course expected to take over at high densities. A fairly complete theoretical treatment of the problem was given in 1961 by Galatry.[3] The main approximations which he made were: 1) that the broadening effect of collisions could be described by phase shifts in the internal state wave functions, and 2) that the correlations in the time and magnitude of collisional phase shifts with the time and magnitude of velocity changes could be ignored. For the case of many collisions during a time equal to the inverse linewidth, he obtained the expected form of dependence on buffer gas density. The predicted line width is the sum of two terms, one inversely proportional to the density and the other directly proportional to the density.

The experimental results were obtained at 300°K using samples about 10 cm in diameter. A correction of about 1.5 Hz was made for Rb–Rb collisions. For He and Ne the densities used corresponded to pressures of between 10 and 375 Torr and between 3 and 400 Torr respectively. The results agreed quite well with the theory, with the minimum line width coming at roughly 30 Torr in both cases. The minimum widths were about 7 Hz for Ne and 10 Hz for He.

For Ar, N_2, and CH_4 as buffer gases, the results do not agree with the theory. If the coefficients of the two terms in the expression for the line width are chosen to fit the highest and lowest density points, then the predicted linewidths at intermediate densities are much less than the observed values. For Ar and CH_4 the discrepancies are about 15 Hz at a density corresponding to 10 Torr. In view of the good agreement and narrow line widths obtained for He and Ne, the effect cannot be an instrumental one. The discrepancies could be explained away by assuming that the 10 Torr samples of Ar and N_2 plus several containing CH_4 were contaminated, but this does not seem likely. The frequencies for these samples agree well with the expected frequencies, and there is little reason for the intermediate density samples to be the only ones affected. Nitrogen is the main impurity expected in argon, and it is itself a good buffer gas. Oxygen is gettered by rubidium, and any residual density which was not gettered would presumably cause broadening in the He and Ne samples also. It seems unlikely that differences in the initial oxygen impurity content would be important, since samples filled initially with air give good signals after the gettering process is completed.

Instead, it appears that the extra hyperfine transition line width may be related to the anomalous Zeeman transition relaxation times observed by Aymar, Bouchiat, Brossel, and Pottier[4,5] for Rb in a Kr buffer gas. The long correlation times of the order of 10^{-8} sec which were obtained appear to be caused by temporary Rb–Kr molecule formation.[6] This effect has also been observed for Rb in Ar and Xe.[5]

For hyperfine transitions, the fractional shift in frequency during the molecular lifetime would probably be large. To be specific, we consider the case of Ar as the buffer gas. If Rb–Ar molecule formation is caused mainly by three-body collisions, the resulting contribution to the linewidth would be expected to increase quadratically with Ar density at low densities. At higher densities, collisions of the Rb–Ar molecule with a free Ar atom would destroy the molecule before the phase shift δ of the internal state wave function due to the molecule formation became large. Since the line broadening is proportional to $\overline{\delta^2}$ for small values of δ, and since δ should be proportional to the molecular lifetime and thus inversely proportional to density, this loss in broadening effectiveness at higher pressures should counter-act the density-squared dependence of the molecular formation rate. We have assumed here that the part of the phase shift due to the collisions which form and destroy the molecule is small compared with the part which accumulates during the molecular lifetime. The resulting contribution to the line width from molecular formation may thus approach a constant value over some range of densities, and could explain the discrepancy between the experimental linewidth results and Galatry's.

References

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