Fluctuating Local Magnetic Moments in Ferromagnetic Ni Observed by the Spin-Resolved Resonant Photoemission

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We have measured the spin- and angle-resolved resonant photoemission spectra of ferromagnetic Ni(110) near the $3p$ threshold in the temperature range from $T/T_C=0.5$ to 1.05. The spin polarization of the 6 eV satellite decreases with increasing temperature. The photon energy dependence of the spin polarization of the 6 eV satellite shows a dip profile near the $3p$ threshold, while its spectral feature remains unaltered at elevated temperature. These results indicate the evidence of the fluctuating local magnetic moments in ferromagnetic Ni.

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The electronic structures of the ferromagnetic $3d$ metals (Fe,Co,Ni) have been a subject of intensive research for a long time [1]. The electronic and magnetic ground state properties of the ferromagnetic $3d$ metals are reasonably understood by the Stoner model of the itinerant electron ferromagnetism which is based on the mean field approximation for the electron-electron interaction and neglects the spin fluctuation. While the Stoner model is consistent with the ground state properties such as the nonintegral number of Bohr magnetons per atom, etc., the finite temperature properties of the ferromagnetic $3d$ metals are not fully explained by the Stoner model. The Curie temperature (T_C) expected by the model is higher than the observed one [2] and the existence of the short range magnetic order above T_C observed by the neutron scattering [3] does not agree with the Stoner model. In the Stoner model, the exchange splitting between spin-up (majority spin) and spin-down (minority spin) valence states decreases with increasing temperature proportional to the macroscopic magnetization and vanishes (collapses) at T_C. Hence the local magnetic order vanishes above T_C.

It has been ten years since Hopster, Raue, and Güntherodt [4] reported the spin- and angle-resolved photoemission of Ni at temperature close to T_C. This experiment motivated the developments of the spin-resolved photoemission studies of the magnetic phase transition of the ferromagnetic $3d$ metals. In spin- and angle-resolved photoemission spectra of Ni [4,5], the exchange splitting in the valence band reduces (collapses) to zero as the temperature increases to T_C, which suggests the validity of the Stoner model of the ferromagnetic phase transition. On the other hand, in spin-resolved valence band photoemission of Fe [6,7], temperature independent peaks were observed. The spin polarization of each peak decreases as temperature increases to T_C, and the collapsing behavior of the exchange split valence bands is not likely to occur. Theoretical interpretations of the spin-resolved valence band photoemission of Fe have been proposed based on the so-called fluctuating local magnetic moment model [8,9].

The temperature dependence of the exchange splitting of the valence bands in Ni was also investigated by means of the spin dependent inelastic electron scattering [10] and the spin polarized electron energy loss spectroscopy [11]. In those experiments, the exchange splitting is not likely temperature dependent. In the inverse photoemission experiment [12], collapsing behavior of the valence bands was observed. Korenman and Prange [13] showed that the collapsing and noncollapsing behavior of the exchange split bands in Ni could be explained qualitatively on the basis of the local band theory [14]. They emphasized that the difference of the group velocities of the photoexcited valence holes played an important role. A valence hole with a small group velocity sees the local exchange field and its spin follows the local magnetization direction, which corresponds to noncollapsing behavior. On the other hand, a valence hole with a large group velocity sees many local exchange fields and its spin follows the average magnetization direction, which corresponds to the collapsing behavior. So far, experimental investigations were only performed in several restricted systems and the quantitative comparison between experiments and theoretical calculations for realistic band structures of Ni is not quite satisfactory. It is not fully understood whether the collapsing behavior of Ni valence bands is due to the reduction of the exchange splitting or not. To clarify the temperature dependence of the valence bands of the ferromagnetic $3d$ metals, experimental evidences obtained by a different and independent technique are needed.

In this Letter, we investigate the temperature dependence of the $3p$-$3d$ resonance aspects of the spin- and angle-resolved resonant photoemission spectra of the 6 eV

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satellite of Ni(110) and show the evidence of the fluctuating local magnetic moments in Ni.

Experiments were carried out at the Revolver undulator beam line BL-19A of the Photon Factory and by utilizing the simultaneous scanning of the magnet gap of the undulator and the output photon energy of the monochromator [15]. The beam line is equipped with an angle-resolved photoelectron spectrometer, a sample preparation chamber, and a Mott detector for the electron spin analysis. The Ni(110) sample was shaped into a picture frame so that each frame was oriented along the easy magnetization axis of ⟨111⟩. The photoelectrons emitted normal to the sample surface were collected. A clean Ni(110) surface was prepared by repeated cycles of ion bombardment and annealing. The cleanliness of the sample surface was checked by LEED and Auger electron spectroscopy before and after each photoemission measurement. The sample was heated by an electron bombardment. The stability of the temperature during the measurement was within $\pm 2°C$. The pressure during the measurements was lower than 1.5×10^{-10} Torr. The energy resolution of the photoemission spectra was 0.25 eV at an excitation energy of 20 eV and 0.6 eV at 70 eV. Since the spin polarization of the 6 eV satellite decreases with increasing temperature, the measuring time at elevated temperature was substantially increased to obtain the spin polarization with small statistical errors. A more detailed description of the experiment is presented elsewhere [16].

Figure 1 shows the temperature dependence of the valence band photoemission spectra of a ferromagnetic Ni(110) near the $3p$ threshold (67 eV), where the valence band satellite at the binding energy of about 6 eV is resonantly enhanced. The spectral features in the main valence band change slightly as a function of temperature due to the temperature dependence of the spectral features in both the majority and minority spin states. This was confirmed by the spin-resolved photoemission spectra [17]. The spectral feature of the 6 eV satellite does not show appreciable temperature dependence.

There have been many theoretical and experimental studies on the photoelectron spectroscopy of the Ni valence band and its 6 eV satellite [18–22]. The origin of the enhancement of the 6 eV satellite near the $3p$ threshold is now understood as being due to the interference effect between the direct $3d$ valence electron excitation ($3p^63d^9 \rightarrow 3p^63d^8+el$) and the $3p$ core electron excitation following by the $M_{2,3}VV$ super-Coster-Kronig (sCK) transition forming the two $3d$ holes in a same atomic site ($3p^63d^9 \rightarrow 3p^53d^{10} \rightarrow 3p^63d^8+el$). The final state of the resonant photoemission mainly consists of the spin singlet ($^1S, ^1G$) and triplet ($^3P, ^3F$) $3d^8$ configurations and the dominant contribution to the 6 eV satellite is due to the ^1G term. Near the $3p$ threshold, the spin singlet ^1G term is achieved by photoexcitation of the $3p$ electrons to the unoccupied $3d$ minority spin states above the Fermi level producing the minority spin $3p$ hole and following sCK decay into the spin singlet final state. It has been assumed here that the majority spin states are occupied in the ground state.

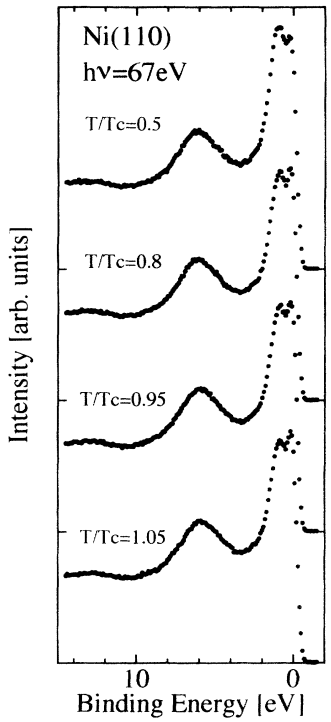

FIG. 1. Photoemission spectra of ferromagnetic Ni(110) near the $3p$ threshold (67 eV) in the temperature range from $T/T_C=0.5$ to 1.05. Photoelectrons emitted normal to the surface were collected. The spectral feature of the 6 eV satellite, which is resonantly enhanced at all temperatures, does not show an appreciable temperature dependence, whereas the spectral features of the main valence band change slightly with temperature.

The temperature independent spectral profiles of the 6 eV satellite in Fig. 1 suggest that the photoelectron excitation process that forms the two $3d$ holes is unaltered at elevated temperature. If the exchange splitting of the valence band of Ni decreases with increasing temperature as expected in the Stoner model, the $3d^8$ configuration of the final state is achieved by the $3p$ electron excitation to both unoccupied minority and majority spin states and following sCK decay. Hence the weight of the spin singlet term in the $3d^8$ final state configuration changes as temperature increases and the spectral feature of the 6 eV satellite would show a temperature dependence.

Figure 2 shows the photon energy dependence of the spin polarization of the 6 eV satellite at various temperatures. We adopted the spectral feature at a binding energy of 6.2 eV for photon energy dependence of the spin polarization of the 6 eV satellite, since the ^1G term in the atomic model is known to be situated around 6.2 eV within the experimental energy resolution [16,21]. The spin polarization of the 6 eV satellite in Fig. 2 decreases with increasing temperature, while the photon energy

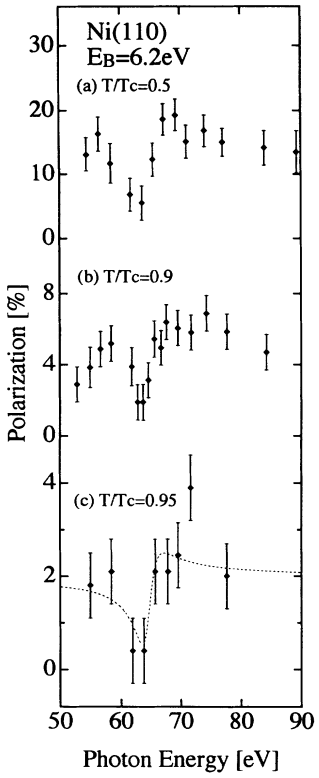

FIG. 2. Photon energy dependence of the spin polarization of the 6 eV satellite at various temperatures: (a) $T/T_C=0.5$, (b) $T/T_C=0.90$, and (c) $T/T_C=0.95$. The empirically determined Fano profile, with $q=0.6$ and $h\nu_0=64.5$ eV, is shown by a thin dotted curve in (c).

dependence at each temperature has common aspects: a strong dip profile near the $3p$ threshold and gradual decreases to both smaller and larger photon energy regions. Although the dip profile in the spectra could not be simply described by the Fano profile, we adopted the Fano profile to represent the spectral profile in Fig. 2 semiquantitatively. The remarkable point is that all three spectra in Fig. 2 could be fitted roughly by the same parameters: the strength of the dip profile, $q=0.6$, and the threshold photon energy, $h\nu_0=64.5$ eV. The results are shown in Fig. 2(c) as a thin dotted line.

So far, the spin polarization of the 6 eV satellite has been studied theoretically on the basis of the atomic model and expected to take its maximum value near the $3p$ threshold [19,20]. Experimentally, it was found that the spin polarization of the 6 eV satellite takes its maximum value of about 15% at room temperature without background correction and its photon energy dependence shows a dip profile near the $3p$ threshold [16]. Recently, Tanaka and Jo [23] interpreted the photon energy dependence of the spin polarization of the 6 eV satellite based on the hybridized $3d$ orbital model where the solid state effects are adopted as the hybridization between $3d$ states and other valence states. They came to the conclusion

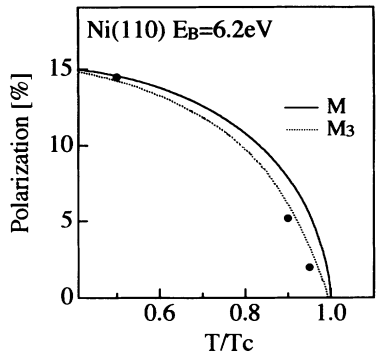

FIG. 3. Temperature dependence of the height of the dip profile in the spin polarization of the 6 eV satellite. The solid and dotted curves represent the temperature dependencies of the macroscopic magnetization (M) and the magnetization of 3 atomic monolayers in the mean field theory (M_3), respectively. The height of the dip profile is normalized to the macroscopic magnetization at $T/T_C=0.5$.

that at low temperature, the dip profile in the spin polarization near the $3p$ threshold is mainly due to the photon energy dependence of the ^1G term in the final state of the photoemission. According to the model calculation [23], the spectral profile of the spin polarization strongly depends on the weight of each component term (^1S, ^1G, ^3P, ^3F) in the $3d^8$ final state configuration. If the exchange splitting of the valence band of Ni changes with increase of temperature, the spectral profile would be altered. The temperature independent line shape of the dip structure in Fig. 2 implies that the photoelectron excitation process to the $3d^8$ final state configuration does not depend on temperature and that the decrease of the spin polarization occurs after photoelectron excitation.

Based on our analysis on the local band theory, we assume that the local magnetization is temperature independent and its direction fluctuates around the macroscopic magnetization direction in a time scale (10^{-12} -10^{-13} sec) much longer than the photoelectron excitation process (10^{-14}–10^{-15} sec). Since we extract photoelectrons at finite temperatures through the fast photoexcitation process, an observed photoelectron reflects the instantaneous local electronic structures at a point in the crystal where the direction of the local magnetization is tilted with respect to the macroscopic magnetization direction. As the spin-resolved photoemission experiment is time integrating and the integrating time (~ 10 sec) is infinitely longer than the spin fluctuating time, we have observed an average magnetization over many tilted local magnetization. Hence the spin polarization of the 6 eV satellite observed in the spin-resolved resonant photoemission would decrease with increasing temperature and trace the same temperature dependence as the macroscopic magnetization.

In Fig. 3, we have plotted the height in the dip profiles shown in Fig. 2 as a function of temperature together

with the macroscopic magnetization (M). The height is normalized to the macroscopic magnetization at $T/T_C = 0.5$. The height of the spin polarization in the dip profile decreases with increasing temperature and shows a quite similar temperature dependence to the macroscopic magnetization. The disappearance of the dip profile at T_C corresponds to the nonexistence of the average magnetic moment along the macroscopic magnetization direction, i.e., the disappearance of the long range magnetic order above T_C. Actually, the spin polarization in the dip profile decreases slightly faster than the magnetization of the 3 atomic monolayers (M_3). This is consistent to the estimated mean probing depth of the photoelectrons. For the 6 eV satellite of Ni at an excitation energy of 67 eV, the probing depth is estimated to be 2 atomic monolayers.

In conclusion, the photon energy dependence of the spin polarization of the 6 eV satellite is well explained by the existence of the temperature independent local magnetization which fluctuates at elevated temperature. This confirms the existence of the temperature independent exchange splitting in the local ferromagnetic bands of Ni.

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